

# Semiclassical kinetic theory of electron spin relaxation in semiconductors

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We develop a semiclassical kinetic theory for electron spin relaxation in semiconductors. Our approach accounts for elastic as well as inelastic scattering and treats Elliott-Yafet and motional-narrowing processes, such as D'yakonov-Perel' and variable  $g$ -factor processes, on an equal footing. Focusing on small spin polarizations and small momentum transfer scattering, we derive, starting from the full quantum kinetic equations, a Fokker-Planck equation for the electron spin polarization. We then construct, using a rigorous multiple time scale approach, a Bloch equation for the macroscopic ( $\vec{k}$ -averaged) spin polarization on the long time scale, where the spin polarization decays. Spin-conserving energy relaxation and diffusion, which occur on a fast time scale, after the initial spin polarization has been injected, are incorporated and shown to give rise to a weight function which defines the energy averages required for the calculation of the spin relaxation tensor in the Bloch equation. Our approach provides an intuitive way to conceptualize the dynamics of the spin polarization in terms of a "test" spin polarization which scatters off "field" particles (electrons, impurities, phonons). To illustrate our approach, we calculate for a quantum well the spin lifetime at temperatures and densities where electron-electron and electron-impurity scattering dominate. The spin lifetimes are non-monotonic functions of temperature and density. Our results show that at electron densities and temperatures, where the cross-over from the non-degenerate to the degenerate regime occurs, spin lifetimes are particularly long.

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## I. INTRODUCTION

The spin degree of freedom of an electron provides an additional variable that potentially can be used to add new functionality to electronic, optoelectronic and magnetoelectronic devices or to even build radically new devices entirely based on the coherence of electron spin states. This has led to the newly emerging field of spintronics. [1, 2] A subclass of spintronics device concepts relies on the capability to inject, control, and detect electron spin polarizations in non-magnetic semiconductors. [3, 4] The spin polarization, which would enable the device operation, is a non-equilibrium state and its characterization, e.g., in terms of lifetimes and transport coefficients, has to be given within a kinetic theory.

Of particular interest is the lifetime of the non-equilibrium spin polarization in non-magnetic n-type III-V semiconductors. Important spin relaxation processes for itinerant electrons in this class of materials include the Elliott-Yafet (EY) process [5, 6], that leads to spin-flip scattering and, in materials without inversion symmetry, the D'yakonov-Perel' (DP) process [7] in which spin states precess because of spin off-diagonal Hamiltonian matrix elements resulting from a combination of spin-orbit coupling and inversion asymmetry. An external magnetic field, in many cases required to control and manipulate the electron spin, can also influence the electron spin dynamics. It quenches the DP process [8], thereby tending to extend the spin lifetimes as a function of magnetic field, and it opens a spin relaxation channel due to the  $\vec{k}$ -dependence of the electron  $g$ -factor, which forces the spin of electrons in different quantum states to precess around an external magnetic field with different rates. [9, 10] For brevity we will refer to this mechanism as a variable  $g$ -factor (VG) process.

Spin dynamics in semiconductors has been extensively studied in magneto-optics [11, 12, 13, 14] using various spin-sensitive emission, transmission, and reflection spectroscopies. These spectroscopies are now readily adaptable to spatially and time resolved measurements [15, 16, 17, 18, 19, 20, 21, 22, 23, 24, 25, 26, 27, 28, 29, 30, 31, 32, 33] which, together with the emergence of spintronics concepts, inspired new theoretical investigations in bulk [10, 34, 35, 36, 37, 38] and dimension-reduced semiconductors. [39, 40, 41, 42, 43, 44, 45, 46, 47, 48, 49, 50, 51, 52, 53]

The theoretical investigations are based on the early work [5, 6, 7] augmented by modern band structure theory for bulk and dimension-reduced semiconductors. The EY spin relaxation rates are usually calculated using the Golden rule for spin-flip scattering, whereas the spin-flip rates due to motional-narrowing (DP and VG) processes are, at least conceptually, obtained from a semiclassical Boltzmann-type equation for the non-equilibrium spin polarization, although not always is the Boltzmann equation explicitly solved. Instead, a common procedure is to adapt the expression for the spin relaxation rate originally derived by D'yakonov and Perel' [7] to the scattering processes under consideration.

The D'yakonov-Perel' expression for the spin relaxation rate, which results from the solution of the Boltzmann

equation within the elastic approximation, treats all scattering processes on-shell, even inelastic scattering events, e.g., due to electron-electron or electron-phonon scattering. The obtained spin relaxation rates are therefore on-shell rates, which have to be averaged over energy with an appropriate weight function before a comparison with experiments can be attempted. Usually, the difference of the distribution functions for spin-up and spin-down electrons divided by the total number of electrons contributing to the initial spin polarization is used as a weight function. [7, 40, 43] The ad-hoc energy averaging, which is necessary because of the incomplete treatment of inelastic scattering processes, can lead to substantial deviations from the spin relaxation rates obtained, e.g., from a full numerical solution of the Boltzmann equation. [35]

We develop in this paper a systematic kinetic theory for electron spin relaxation, applicable to spin-flip (EY) and motional-narrowing (DP and VG) spin relaxation processes in bulk and quantum wells, which avoids the ad-hoc energy averaging and gives a clear physical picture of the time evolution of the optically or electrically injected non-equilibrium spin polarization. We derive, starting from the full quantum kinetic equations for the electron Green functions, a semiclassical Fokker-Planck equation for the time evolution of the non-equilibrium spin polarization, valid for small spin polarizations and for small momentum transfer scattering, and employ a multiple time scale perturbation approach to separate the fast spin-conserving from the slow spin non-conserving time evolution. As a result, we obtain on the time scale of spin relaxation a Bloch equation for the macroscopic ( $\vec{k}$ -averaged) non-equilibrium spin polarization, which is the quantity measured in, e.g., time-resolved Faraday and Kerr rotation experiments. [16, 17, 18, 19, 29, 30] The weight function defining the energy averages needed, e.g., for the calculation of the spin relaxation tensor and the spin relaxation rates, turns out to be directly related to the quasi-stationary spin polarization, which is the terminating state of the initial, fast spin-conserving time evolution of the injected spin polarization. Our approach treats spin-flip (EY) and motional-narrowing (DP and VG) processes on an equal footing. Due to the different angle dependences, a Matthiessen-type rule holds, however, for isotropic semiconductors, where the total spin relaxation tensor is simply the sum of the individual spin relaxation tensors. [10] The diagonal elements of the spin relaxation tensor, the spin relaxation rates, are either given in terms of an energy averaged spin-flip rate (EY process) or an energy average of a generalized relaxation time, which accounts for both on- and off-shell scattering events, multiplied by a precession rate (DP and VG processes).

In the next section we introduce a generic model for electrons in n-type III-V semiconductors applicable to bulk and quantum well situations. In Sec. III we give a complete description of our semiclassical kinetic theory for the electron spin dynamics. As far as the formal development is concerned, we treat EY, DP and VG processes on an equal footing and also allow for quenching effects due to orbital motion of electrons in an external magnetic field. In Sec. IV we apply our approach to the particular situation of DP spin relaxation in an idealized quantum well at temperatures and densities for which electron-electron and electron-impurity scattering dominate. Our main findings are summarized in Sec. V. Technical details concerning the calculation of the quantum well collision integrals due to electron-electron and electron-impurity scattering are relegated to an Appendix.

## II. MODEL HAMILTONIAN

We consider conduction band (CB) electrons in III-V semiconductors, e.g., GaAs, in the presence of an applied magnetic field. The model used here applies to both bulk and quantum well situations. Within an envelope function approach [54, 55], which treats the two states at the conduction band minimum explicitly and includes a large set of states perturbatively, the effective mass Hamiltonian for the CB electrons can be cast into the form

$$H_{\alpha\alpha'}(\vec{K}) = \epsilon(\vec{K})\delta_{\alpha\alpha'} + \frac{\hbar}{2}\vec{\Omega}_L \cdot \vec{\sigma}_{\alpha\alpha'} + \frac{\hbar}{2}[\vec{\Omega}_{IA}(\vec{K}) + \vec{\Omega}_g(\vec{K})] \cdot \vec{\sigma}_{\alpha\alpha'}, \quad (1)$$

where  $\vec{K} = \vec{k} - (e/\hbar c)\vec{A}(\vec{r})$  and  $\vec{A}(\vec{r})$  is the vector potential. The “spin basis” for the CB electrons used to define the model (1) is  $\alpha = +$  and  $\alpha = -$ , where  $\alpha = +$  ( $\alpha = -$ ) denotes a state which is mostly spin-up (spin-down) with a small admixture of spin-down (spin-up).

The first term denotes the dispersion of the Kramers degenerate conduction band which, depending on the sophistication of the envelope function approach, could contain nonparabolicity effects. For quantum wells  $\vec{k}$  and  $\epsilon(\vec{k})$  denote the in-plane momentum and the in-plane dispersion of the conduction subband under consideration. The second term comprises the Larmor precession due to the external magnetic field, with  $\hbar\vec{\Omega}_L = \mu_B g^* \vec{B}$  the Larmor energy vector. Here  $\mu_B$  and  $g^*$  denote the Bohr magneton and the electron  $g$ -factor. The third term describes spin off-diagonal Hamiltonian matrix elements arising from the coupling to higher lying states. The most important of which are the splitting of the conduction band due to inversion asymmetry (IA) and the term which leads to a  $\vec{k}$ -dependent electron

g-factor. For bulk semiconductors, the two contributions are given by

$$\hbar\vec{\Omega}_{IA}(\vec{K}) = 2\delta_0\vec{\kappa}_{IA}(\vec{K}) , \quad (2)$$

$$\hbar\vec{\Omega}_g(\vec{K}) = 2a_4K^2\vec{B} + 2a_5\{\vec{K}, \vec{B} \cdot \vec{K}\} + 2a_6\vec{\tau}(\vec{K}, \vec{B}) , \quad (3)$$

respectively. The definition of the vectors  $\vec{\kappa}_{IA}(\vec{K})$  and  $\vec{\tau}(\vec{K}, \vec{B})$  and of the parameters  $\delta_0$  and  $a_i$  can be found in Refs. [54, 55] and  $\{\dots\}$  denotes an anticommutator. The expressions for conduction subbands in a quantum well are obtained by averaging the bulk expressions (2) and (3) over the subband envelope function.

In addition to bulk inversion asymmetry, dimension-reduced semiconductors can have additional sources of asymmetry due to interfaces which share no common atom [31] or due to layer design (structural inversion asymmetry [39]). Both mechanisms can be cast into spin off-diagonal Hamiltonian matrix elements and can be therefore treated in the same way as the spin off-diagonal terms due to bulk inversion asymmetry.

For a complete description, a collision term arising from electron-impurity, electron-phonon and electron-electron scattering,

$$H_c = H_{ei} + H_{ep} + H_{ee} ,$$

is added to the effective mass Hamiltonian. The electron-impurity term reads

$$H_{ei} = \sum_{\vec{k}\vec{k}'} \sum_{\alpha\alpha'} M_{\alpha,\alpha'}(\vec{k}, \vec{k}') c_{\vec{k}\alpha}^\dagger c_{\vec{k}'\alpha'} , \quad (4)$$

with a scattering matrix element given by

$$M_{\alpha,\alpha'}(\vec{k}, \vec{k}') = \sum_j U(\vec{k} - \vec{k}') e^{i(\vec{k}-\vec{k}')\vec{r}_j} I_{\alpha\alpha'}(\vec{k}, \vec{k}') . \quad (5)$$

The Bloch states for the conduction band are not pure spin states, because of spin-orbit coupling. The scattering matrix element contains therefore an overlap factor

$$I_{\alpha\alpha'}(\vec{k}, \vec{k}') = \langle U_{\alpha,\vec{k}} | U_{\alpha',\vec{k}'} \rangle , \quad (6)$$

which is of order unity for  $\alpha = \alpha'$  (spin conserving scattering) and is small, but not zero otherwise (spin non-conserving scattering). The electron-phonon collision term would have the same structure as Eq. (4) but with phonon creation and annihilation operators appearing in the matrix element  $M_{\alpha\alpha'}(\vec{k}, \vec{k}')$ . The electron-electron scattering contribution has the form

$$H_{ee} = \frac{1}{2} \sum_{\vec{k}_i\alpha_i} M_{\alpha_1\alpha_2\alpha_3\alpha_4}(\vec{k}_1, \vec{k}_2, \vec{k}_3, \vec{k}_4) c_{\vec{k}_1\alpha_1}^\dagger c_{\vec{k}_2\alpha_2}^\dagger c_{\vec{k}_3\alpha_3} c_{\vec{k}_4\alpha_4} , \quad (7)$$

where the scattering matrix element,

$$M_{\alpha_1\alpha_2\alpha_3\alpha_4}(\vec{k}_1, \vec{k}_2, \vec{k}_3, \vec{k}_4) = V(\vec{k}_1 - \vec{k}_4) I_{\alpha_1\alpha_4}(\vec{k}_1, \vec{k}_4) I_{\alpha_2\alpha_3}(\vec{k}_2, \vec{k}_3) \delta_{\vec{k}_1+\vec{k}_2, \vec{k}_3+\vec{k}_4} , \quad (8)$$

contains two overlap factors. The functions  $U(\vec{k})$  and  $V(\vec{k})$  denote, respectively, the potential of a single impurity (neutral or ionized) and the Coulomb potential between two conduction electrons.

The model Hamiltonian is characterized by  $\epsilon(\vec{k})$ ,  $\vec{\Omega}_{IA}(\vec{k})$ ,  $\vec{\Omega}_g(\vec{k})$ , and  $I_{\alpha,\alpha'}(\vec{k}, \vec{k}')$ . These quantities need to be obtained by an electronic structure calculation. The formal structure of the kinetic theory described in the next section is independent of the particular form of these quantities.

### III. SEMICLASSICAL KINETIC THEORY

In this section we give a systematic derivation of the Fokker-Planck equation governing the electron spin relaxation in the limit of small spin polarizations. The derivation is independent of dimensionality, applying to bulk semiconductors and semiconductor heterostructures, and treats motional-narrowing (DP and VG) and spin-flip (EY) spin relaxation processes on an equal footing. To obtain a Fokker-Planck equation, we restrict ourselves to the Born approximation, but collective effects giving rise to dynamical screening of the Coulomb interaction can be approximately incorporated

at the level of a quantum analog to the Lenard-Balescu equation. [56] Besides its intuitive interpretation in terms of a small “test” spin polarization scattering off a bath of “field” particles (impurities, electrons and phonons), causing dynamical friction, diffusion, and eventually relaxation for the “test” spin polarization, the Fokker-Planck equation is the starting point for a multiple time scale analysis which results in the derivation of a Bloch equation for the macroscopic ( $\vec{k}$ -averaged) spin polarization. Its decay is usually characterized by the diagonal elements of a spin relaxation tensor, which are quadratures of either a spin-flip rate (EY process) or a generalized relaxation time multiplied by a precession rate (DP and VG processes).

Since the derivation is quite lengthy and to some extent rather formal we first give a short outline of the main steps. We start from the full quantum kinetic equations for the Keldysh Green functions. [57, 58, 59, 60] Each component of the Keldysh Green function is a  $2 \times 2$  matrix in electron spin space. In the first step we derive, within the semiclassical approximation, a kinetic equation for the density matrix. This amounts to treating momentum scattering processes as instantaneous on the time scale of spin relaxation, which is usually the case. Calculating the self-energies which appear in the semiclassical kinetic equation in the Born approximation, linearizing with respect to spin polarization, and expanding the self-energies up to second order in the momentum transfer (diffusion approximation) finally yields a Fokker-Planck equation for the spin polarization, which we then analyze in terms of multiple time scale perturbation theory.

### A. Kinetic equations

For a spatially homogeneous system (we assume a constant magnetic field  $\vec{B}$ ), the information about spin relaxation is contained in the electronic density matrix, which, due to the spin degree of freedom, is a  $2 \times 2$  matrix in spin space,

$$N_{\alpha_1\alpha_2}(\vec{k}, t) = \langle (c_{\vec{k}\alpha_1}^\dagger c_{\vec{k}\alpha_2})(t) \rangle, \quad (9)$$

but diagonal in  $\vec{k}$ -space. Here, the operators evolve in time with the full Hamiltonian, including the time-dependent perturbation, which could be, e.g., a circularly polarized light pulse applied at time  $t = t_0$ . To perform the averaging in Eq. (9) denoted by  $\langle \dots \rangle$ , we consider the system to be in thermodynamical equilibrium for  $t < t_0$ , take the limit  $t_0 \rightarrow -\infty$  and evaluate the expectation value in Eq. (9) with respect to the equilibrium density matrix. [60]

To derive a kinetic equation for the density matrix it is convenient to start from Keldysh Green functions. [57, 58] For a constant magnetic field, the vector potential is a function of  $\vec{r}$ . It is therefore necessary to initially work with kinetic equations in real space. In this subsection we set  $\hbar = 1$ . Introducing a numerical index 1 that stands for  $\vec{r}_1\alpha_1t_1$  and 2 for  $\vec{r}_2\alpha_2t_2$ , we write in the notation of Ref. [59]

$$i\hat{G}_{12} = i \begin{pmatrix} G_{12}^{++} & G_{12}^{+-} \\ G_{12}^{-+} & G_{12}^{--} \end{pmatrix}. \quad (10)$$

Note that each component of the Keldysh Green function is a  $2 \times 2$  matrix in spin space. Introducing further a self-energy

$$\hat{\Sigma}_{12} = \begin{pmatrix} \Sigma_{12}^{++} & \Sigma_{12}^{+-} \\ \Sigma_{12}^{-+} & \Sigma_{12}^{--} \end{pmatrix}, \quad (11)$$

we set up two matrix Dyson equations, one where the time differentiation is with respect to  $t_1$  and one where it is with respect to  $t_2$ :

$$\partial_{t_1} \hat{G}_{12} = -i\hat{\tau}_z \delta_{12} - i(\hat{\epsilon}\hat{G})_{12} - i\hat{\tau}_z(\hat{\Sigma}\hat{G})_{12}, \quad (12)$$

$$\partial_{t_2} \hat{G}_{12} = i\hat{\tau}_z \delta_{12} + i(\hat{G}\hat{\epsilon})_{12} + i(\hat{G}\hat{\Sigma})_{12}\hat{\tau}_z, \quad (13)$$

with  $\delta_{12} = \delta(t_1 - t_2)\delta(12)$  and the energy matrix  $\hat{\epsilon}_{12} = \delta(t_1 - t_2)\hat{\epsilon}(12) = \delta(t_1 - t_2)\hat{\tau}_0\delta(12)\epsilon(-i\nabla_{\vec{r}_1} - (e/c)\vec{A}(\vec{r}_1))$ , where we neglect nonparabolicities in the dispersion. We adopt the convention that numerical indices written as a subscript contain the time variable, whereas numerical indices written as an argument do not. Matrix multiplication with respect to the Keldysh indices is implied and internal variables are summed (integrated) over;  $\hat{\tau}_z$  is a Pauli matrix and  $\hat{\tau}_0$  is the unit matrix in Keldysh space.

Subtracting Eq. (13) from Eq. (12) gives

$$[\hat{L}, \hat{G}]_{12} = \hat{\tau}_z(\hat{\Sigma}\hat{G})_{12} - (\hat{G}\hat{\Sigma})_{12}\hat{\tau}_z, \quad (14)$$

where  $[\dots]$  denotes the commutator. To compactify the notation, we introduced a differential operator

$$\hat{L}_{13} = \hat{\tau}_0 \delta_{13} L(3) = \hat{\tau}_0 \delta_{13} (i\partial_{t_3} - \epsilon(3)) . \quad (15)$$

It is understood that in the second term of the commutator, the operator  $\hat{L}_{32}$  acts to the left with the temporal differential operator  $\partial_{t_3}$  replaced by its adjoint  $-\partial_{t_3}$ .

Equation (14) contains two time variables. To obtain a kinetic equation for the electronic density matrix, which depends only on a single time variable, it is necessary to perform the equal time limit. This is most conveniently done in the (mixed) Wigner representation, where the equal time limit reduces to an integration. Separating the self-energy into a singular and a regular part [59]

$$\Sigma_{12}^{pq} = \Delta^{pq}(12; t_1) \delta_{pq} \delta(t_1 - t_2) + \tilde{\Sigma}_{12}^{pq} , \quad (16)$$

introducing relative and center variables,  $\vec{r} = \vec{r}_1 - \vec{r}_2$ ,  $\vec{R} = (\vec{r}_1 + \vec{r}_2)/2$ ,  $\tau = t_1 - t_2$ , and  $T = (t_1 + t_2)/2$ , and defining a Fourier transformation with respect to the relative variables,

$$A(\vec{R}, T, \vec{k}, \omega) = \int_{-\infty}^{\infty} d\tau \int d\vec{r} e^{i\omega\tau - i\vec{k}\vec{r}} A(\vec{R}, T, \vec{r}, \tau) , \quad (17)$$

together with a gradient operator [60]

$$\mathcal{G}^{AB} = \exp \frac{1}{2i} \left[ \partial_T^A \partial_\omega^B - \partial_\omega^A \partial_T^B + \nabla_{\vec{k}}^A \cdot \nabla_{\vec{R}}^B - \nabla_{\vec{R}}^A \cdot \nabla_{\vec{k}}^B \right] , \quad (18)$$

the equal time limit of the  $++$  component of Eq. (14) can be written as

$$D(\vec{R}, T, \vec{k}) = F(\vec{R}, T, \vec{k}) + C(\vec{R}, T, \vec{k}) , \quad (19)$$

with a driving term on the lhs,

$$\begin{aligned} D(\vec{R}, T, \vec{k}) &= \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} (\mathcal{G}^{LG} L(\vec{R}, T, \vec{k}, \omega) G^{++}(\vec{R}, T, \vec{k}, \omega) \\ &\quad - \mathcal{G}^{GL} G^{++}(\vec{R}, T, \vec{k}, \omega) L(\vec{R}, T, \vec{k}, \omega)) , \end{aligned} \quad (20)$$

and a rhs, which contains a molecular field term arising from the singular part of the self-energy,

$$\begin{aligned} F(\vec{R}, T, \vec{k}) &= \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} [\mathcal{G}^{\Delta G} \Delta^{++}(\vec{R}, T, \vec{k}) G^{++}(\vec{R}, T, \vec{k}, \omega) \\ &\quad - \mathcal{G}^{G\Delta} G^{++}(\vec{R}, T, \vec{k}, \omega) \Delta^{++}(\vec{R}, T, \vec{k})] , \end{aligned} \quad (21)$$

and a collision term due to the regular part

$$\begin{aligned} C(\vec{R}, T, \vec{k}) &= \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} (\mathcal{G}^{\tilde{\Sigma}G} \tilde{\Sigma}^{++}(\vec{R}, T, \vec{k}, \omega) G^{++}(\vec{R}, T, \vec{k}, \omega) \\ &\quad - \mathcal{G}^{G\tilde{\Sigma}} G^{++}(\vec{R}, T, \vec{k}, \omega) \tilde{\Sigma}^{++}(\vec{R}, T, \vec{k}, \omega) \\ &\quad + \mathcal{G}^{\tilde{\Sigma}G} \tilde{\Sigma}^{+-}(\vec{R}, T, \vec{k}, \omega) G^{-+}(\vec{R}, T, \vec{k}, \omega) \\ &\quad - \mathcal{G}^{G\tilde{\Sigma}} G^{+-}(\vec{R}, T, \vec{k}, \omega) \tilde{\Sigma}^{-+}(\vec{R}, T, \vec{k}, \omega)) . \end{aligned} \quad (22)$$

The semiclassical approximation amounts to the assumption that the Green functions and self-energies vary slowly on the macroscopic scales,  $T$  and  $\vec{R}$ , respectively. It is therefore sufficient to keep in Eq. (19) only the leading order terms in a gradient expansion. The leading order of the rhs of Eq. (19) is the zeroth order, i.e., Eqs. (21) and (22) with  $\mathcal{G}^{AB} \rightarrow 1$ . The lhs of Eq. (19), however, has to be determined to first order, because the zeroth order vanishes. Using

$$L(\vec{R}, T, \omega, \vec{k}) = \omega + \epsilon(\vec{K}) , \quad (23)$$

we explicitly obtain

$$\begin{aligned} D(\vec{R}, T, \vec{k}) &= i \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} [\partial_T G^{++}(\vec{R}, T, \vec{k}, \omega) - \nabla_{\vec{R}} \epsilon(\vec{K}) \cdot \nabla_{\vec{k}} G^{++}(\vec{R}, T, \vec{k}, \omega) \\ &\quad + \nabla_{\vec{k}} \epsilon(\vec{K}) \cdot \nabla_{\vec{R}} G^{++}(\vec{R}, T, \vec{k}, \omega) . \end{aligned} \quad (24)$$

To ensure gauge invariance of the kinetic equation we follow Ref. [61] and consider the generalized momentum  $\vec{K} = \vec{k} - (e/c)\vec{A}(\vec{R})$  as an independent variable instead of the momentum  $\vec{k}$ . Using the two identities [61],

$$\begin{aligned}\nabla_{\vec{R}} a(\vec{K}) &= \nabla_{\vec{K}} a(\vec{K}) \cdot \nabla_{\vec{R}} \vec{K} + \nabla_{\vec{K}} a(\vec{K}) \times (\nabla_{\vec{R}} \times \vec{K}) , \\ \vec{a} \cdot \vec{b} \times (\nabla_{\vec{R}} \times \vec{c}) &= (\vec{a} \cdot \nabla_{\vec{R}} \vec{c}) \cdot \vec{b} - (\vec{b} \cdot \nabla_{\vec{R}} \vec{c}) \cdot \vec{a} ,\end{aligned}$$

Eq. (24) becomes

$$D(\vec{R}, T, \vec{K}) = \left( \partial_T + \frac{e}{c} \nabla_{\vec{K}} \epsilon(\vec{K}) \cdot (\vec{B} \times \nabla_{\vec{K}}) + \nabla_{\vec{K}} \epsilon(\vec{K}) \cdot \nabla_{\vec{R}} \right) i \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} G^{++}(\vec{R}, T, \vec{K}, \omega) . \quad (25)$$

We assume weak interactions and replace the full Green functions by the noninteracting Green functions (quasi-particle Ansatz),

$$G^{pq}(\vec{R}, T, \vec{K}, \omega) = G_0^{pq}(\vec{k} \rightarrow \vec{K}, \omega)|_{N_0(\vec{k}) \rightarrow N(\vec{R}, T, \vec{K})} , \quad (26)$$

where the noninteracting density matrix  $N_0(\vec{k})$  is replaced by the full density matrix  $N(\vec{R}, T, \vec{K})$ . Performing the  $\omega$ -integrations in Eqs. (21), (22), and (25) then yields the semiclassical kinetic equation for the electronic density matrix:

$$\begin{aligned}\left( \partial_T + \frac{e}{c} \nabla_{\vec{K}} \epsilon(\vec{K}) \cdot \vec{B} \times \nabla_{\vec{K}} + \nabla_{\vec{K}} \epsilon(\vec{K}) \cdot \nabla_{\vec{R}} \right) N(\vec{R}, T, \vec{K}) = \\ i[N(\vec{R}, T, \vec{K}), \Delta^{++}(\vec{R}, T, \vec{K}) + \tilde{\Sigma}^{++}(\vec{R}, T, \vec{K}, \epsilon(\vec{K}))] \\ + iN(\vec{R}, T, \vec{K})\tilde{\Sigma}^{-+}(\vec{R}, T, \vec{K}, \epsilon(\vec{K})) + i\tilde{\Sigma}^{+-}(\vec{R}, T, \vec{K}, \epsilon(\vec{K}))[1 - N(\vec{R}, T, \vec{K})] .\end{aligned} \quad (27)$$

To obtain a closed kinetic equation for the electronic density matrix, internal Green functions, which appear in the self-energies, have to be of course also eliminated according to Eq. (26). Details concerning the calculation of self-energies are given in the next section.

For a homogeneous magnetic field, the electronic density matrix does not explicitly depend on  $\vec{R}$ . The  $\vec{R}$  dependence can be therefore neglected. For a quadratic dispersion,  $\epsilon(\vec{K}) = \vec{K}^2/2m^*$  ( $\hbar = 1$  in this subsection), the Lorentz term moreover becomes

$$(e/c)\nabla_{\vec{K}} \epsilon(\vec{K}) \times \vec{B} \cdot \nabla_{\vec{K}} = (\vec{K} \times \vec{\Omega}_C) \cdot \nabla_{\vec{K}} = -i\vec{\Omega}_C \cdot \vec{\mathcal{L}} , \quad (28)$$

where  $\vec{\Omega}_C = e\vec{B}/m^*c$  is the cyclotron energy vector and  $\vec{\mathcal{L}}$  the angular momentum operator in  $\vec{K}$ -space, and we obtain the kinetic equation for the electronic density matrix in a more familiar form:

$$\begin{aligned}\left( \partial_T - i\vec{\Omega}_C \cdot \vec{\mathcal{L}} \right) N(T, \vec{K}) = i[N(T, \vec{K}), \Delta^{++}(T, \vec{K}) + \tilde{\Sigma}^{++}(T, \vec{K}, \epsilon(\vec{K}))] \\ + iN(T, \vec{K})\tilde{\Sigma}^{-+}(T, \vec{K}, \epsilon(\vec{K})) \\ + i\tilde{\Sigma}^{+-}(T, \vec{K}, \epsilon(\vec{K}))[1 - N(T, \vec{K})] .\end{aligned} \quad (29)$$

This equation is the basis for the calculation of the spin relaxation time in spatially homogeneous systems subject to a constant magnetic field. The first term on the rhs describes the coherent motion in a molecular field modified by correlation effects. If  $\Delta^{++}$ ,  $\tilde{\Sigma}^{++}$ , and  $N$  were scalar functions, as in ordinary transport theory, this term would vanish. The molecular field term is therefore a consequence of the quantum mechanical treatment of the spin degree of freedom. To the singular part of the self-energy contribute the spin off-diagonal terms in the Hamiltonian and the Hartree-Fock fields due to electron-electron scattering. Dissipation and relaxation originate from the regular part of the self-energy and give rise to the second and third term on the rhs. They are at least second order in the interaction. Formally, they correspond to the scattering-out and the scattering-in terms in a matrix-Boltzmann equation. The matrix structure is of course a consequence of the full quantum mechanical description of the spin. Only momentum scattering is treated classically.

## B. Calculation of the self-energies

The semiclassical approach to furnish the self-energies in the matrix-Boltzmann equation (29), valid for magnetic fields, which do not restructure the electron dispersion, is to represent interaction processes in terms of diagrams,

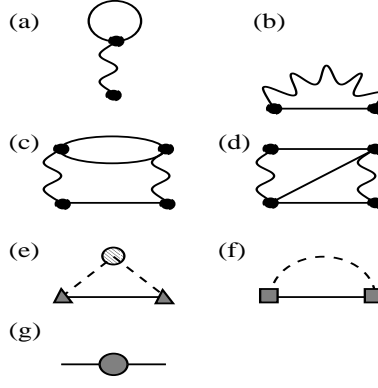


FIG. 1: Diagrammatic representation of self-energies in the Born approximation for electron-electron (a)–(d), electron-impurity (e), and electron-phonon scattering (f). Diagram (g) denotes the self-energy due to spin off-diagonal Hamiltonian matrix elements.

calculate the diagrams using standard rules [58] to obtain  $\tilde{\Sigma}^{pq}(\vec{k}, t, t')$  and  $\Delta^{pq}(\vec{k}, t)$ , perform the zeroth order gradient expansion, and then replace all internal Green functions according to Eq. (26). This heuristic strategy leads to self-energies, which can be expressed in a manifestly gauge invariant form by writing the internal momentum integrations in terms of the generalized momentum  $\vec{K}$ . The formal structure of the self-energies is then the same as without magnetic field. Within the semiclassical approach, the magnetic field gives therefore only rise to the Lorentz term.

In Fig. 1 we depict the self-energies in the Born approximation for electron-electron (a–d), electron-impurity (e), and electron-phonon (f) scattering. Diagram (g) corresponds to the self-energy due to the spin off-diagonal term in the Hamiltonian (1).

The Hartree-Fock diagrams (a) and (b) contribute to the instantaneous self-energy  $\Delta^{++}$ . They are second order in the spin polarization and therefore, for sufficiently small spin polarizations, negligible. There are two second order diagrams due to electron-electron scattering, the direct (c) and the exchange (d) Born diagram. Anticipating that soft scattering dominates, we neglect the exchange diagram (d). The direct Born diagram contributes to  $\tilde{\Sigma}^{++}$ ,  $\tilde{\Sigma}^{+-}$ , and  $\tilde{\Sigma}^{-+}$ . It can be shown, however, that  $\tilde{\Sigma}^{++}$  is at least second order in the spin polarization and therefore negligible in the limit of small spin polarizations. The  $\tilde{\Sigma}^{-+}$  and  $\tilde{\Sigma}^{+-}$  components, contributing to the collision integral, are in contrast linear in the spin polarization and cannot be neglected. Diagrams (e) and (f), corresponding to the Born approximation for electron-impurity and electron-phonon scattering, only contribute to  $\tilde{\Sigma}^{pq}$ . As in the case of electron-electron scattering, the  $++$  component can be again neglected, if the spin polarization is small enough, whereas the  $+-$  and  $-+$  components contribute in linear order in the spin polarization to the collision integral. Diagram (g), corresponding to the spin off-diagonal part of the Hamiltonian (1), is linear in the spin polarization and contributes to  $\Delta^{++}$ . Eventually it leads to a torque force acting on the spin polarization.

Anticipating small spin polarizations, we neglect  $\tilde{\Sigma}^{++}$  and the Hartree-Fock contribution to  $\Delta^{++}$ . Writing in the Born approximation furthermore  $I_B[N] = I_B^{ee}[N] + I_B^{ep}[N] + I_B^i[N]$  for the second and third term on the rhs of Eq. (29), the semiclassical kinetic equation for the electronic density matrix reduces to

$$\left( \hbar \partial_t - i \hbar \vec{\Omega}_C \cdot \vec{\mathcal{L}} \right) N(\vec{k}, t) = \frac{i}{2} \left[ N(\vec{k}, t), \left( \hbar \vec{\Omega}_L + \hbar \vec{\Omega}_{IA}(\vec{k}) + \hbar \vec{\Omega}_g(\vec{k}) \right) \cdot \vec{\sigma} \right] + I_B[N], \quad (30)$$

where we relabeled the center time  $T \rightarrow t$  and adjusted to the notation of Eq. (9);  $\hbar$  is explicitly included and the generalized momentum is now denoted by  $\vec{k}$ .

Equation (30) is a matrix Boltzmann equation similar to the semiconductor Bloch equations frequently used to describe optically pumped semiconductors.[60] Thus, numerical techniques used for the solution of the semiconductor Bloch equations can be adopted to the numerical solution of Eq. (30). Calculations of this kind have been successfully performed for various situations. [34, 35, 44, 51, 52, 53]

To avoid a numerical solution, we focus on small spin polarizations and linearize the Born collision integral with respect to the spin polarization. It is important to note that the equilibrium density matrix  $N_{eq}(\vec{k}) = N(\vec{k}, t \rightarrow \infty)$  is not diagonal in the “spin basis”. Expanding the equilibrium density matrix in terms of Pauli matrices yields

$$N_{eq}(\vec{k}) = f(\vec{k}) + \frac{1}{2} \vec{\sigma} \cdot \vec{S}_{eq}(\vec{k}), \quad (31)$$

where  $f(\vec{k}) = (1/2) \text{Tr} N_{eq}(\vec{k}) = (f_+(\vec{k}) + f_-(\vec{k}))/2$  is half of the sum of the equilibrium distribution functions of the spin up and spin down electrons and  $\vec{S}_{eq}(\vec{k})$  is the equilibrium spin polarization. Accordingly, we also write for the

density matrix at arbitrary times

$$N(\vec{k}, t) = f(\vec{k}) + \delta f(\vec{k}, t) + \frac{1}{2} \vec{\sigma} \cdot [\vec{S}_{eq}(\vec{k}) + \delta \vec{S}(\vec{k}, t)] , \quad (32)$$

with  $\delta f(\vec{k}, t)$  and  $\delta \vec{S}(\vec{k}, t)$  the changes induced by optical pumping or by electrical injection. We defined  $N_{eq}(\vec{k})$  for  $t \rightarrow \infty$ , that is, it contains the electrons created by the perturbation and both  $\delta \vec{S}(\vec{k}, t)$  and  $\delta f(\vec{k}, t)$  have to vanish for  $t \rightarrow \infty$ .

Inserting the expansion (32) into the Boltzmann equation (30) yields two kinetic equations, one for the charge component  $\delta f(\vec{k}, t)$  and one for the spin component  $\delta \vec{S}(\vec{k}, t)$ . The collision terms couple the two equations. If however only a small portion of the total number of electrons initially contributed to the spin polarization, i.e. if  $\delta f(\vec{k}, t) \ll f(\vec{k})$ , the coupling can be ignored and it suffices to focus on the equation for  $\delta \vec{S}(\vec{k}, t)$  alone.

Since the total spin polarization  $\vec{S}_{eq}(\vec{k}) + \delta \vec{S}(\vec{k}, t)$  is small, we linearize the Born collision integral with respect to both  $\vec{S}_{eq}(\vec{k})$  and  $\delta \vec{S}(\vec{k}, t)$ . Thus,

$$I_B[N] = I_B[N_{eq}] + I_B[f + \delta f, \delta \vec{S}] . \quad (33)$$

If we now apply  $\text{Tr} \vec{\sigma}[\dots]$  on both sides of Eq. (30), use  $I_B[N_{eq}] = 0$  as well as  $\vec{\Omega}_{IA}(\vec{k}) \times \vec{S}_{eq}(\vec{k}) = 0$ , because, by construction, the equilibrium density matrix  $N_{eq}(\vec{k})$  commutes with  $H_{IA}$ , and ignore furthermore  $\delta f$  in  $I_B[f + \delta f, \delta \vec{S}]$ , we get a closed kinetic equation for the non-equilibrium spin polarization

$$\left( \hbar \partial_t - i \hbar \vec{\Omega}_C \cdot \vec{\mathcal{L}} \right) \delta \vec{S}(\vec{k}, t) = \left( \hbar \vec{\Omega}_L + \hbar \vec{\Omega}_{IA}(\vec{k}) + \hbar \vec{\Omega}_g(\vec{k}) \right) \times \delta \vec{S}(\vec{k}, t) + J_B[f, \delta \vec{S}] \quad (34)$$

with  $J_B[f, \delta \vec{S}] = \text{Tr} \vec{\sigma} I_B[f, \delta \vec{S}]$ .

The collision integral can be further simplified if we split the spin-flip matrix into a leading spin conserving diagonal part and a small off-diagonal part which describes spin-flip scattering. Since nonparabolicities are small, the diagonal part is approximately equal to the unit matrix and we get  $I(\vec{k}, \vec{k}') \simeq 1 + \delta I(\vec{k}, \vec{k}')$ , with  $\delta I(\vec{k}, \vec{k}') \ll 1$ . Expanding the collision integrals up to second order in  $\delta I(\vec{k}, \vec{k}')$  gives

$$J_B[f, \delta \vec{S}] = J_B[f, \delta \vec{S}]^{(0)} + J_B[f, \delta \vec{S}]^{(1)} + J_B[f, \delta \vec{S}]^{(2)} . \quad (35)$$

The first order term  $J_B^{(1)}[f, \delta \vec{S}]$  potentially mixes EY, DP, and VG spin relaxation channels, but for semiconductors with high symmetry it does not contribute to the spin relaxation rates.

The kinetic equation for the excess spin polarization becomes therefore

$$\begin{aligned} \left( \hbar \partial_t - i \hbar \vec{\Omega}_C \cdot \vec{\mathcal{L}} \right) \delta \vec{S}(\vec{k}, t) &= \hbar \vec{\Omega}_L \times \delta \vec{S}(\vec{k}, t) + \left( \hbar \vec{\Omega}_{IA}(\vec{k}) + \hbar \vec{\Omega}_g(\vec{k}) \right) \times \delta \vec{S}(\vec{k}, t) \\ &+ J_B^{(0)}[f, \delta \vec{S}] + J_B^{(2)}[f, \delta \vec{S}] . \end{aligned} \quad (36)$$

This equation contains motional-narrowing (DP and VG) and spin-flip (EY) spin relaxation processes on an equal footing. The Elliott-Yafet process is simply encoded in  $J_B^{(2)}[f, \delta \vec{S}]$  whereas the motional-narrowing processes result from the combined action of the torque forces given by the second term on the rhs and the spin conserving scattering processes comprising  $J_B^{(0)}[f, \delta \vec{S}]$ .

Independent of the scattering process, the structure of the collision integrals in Eq. (36) is ( $\nu = \text{ei}, \text{ee}, \text{and ep}$ )

$$J_\nu^{(0)}[f, \delta \vec{S}] = \sum_{\vec{q}} [W^\nu(\vec{k} + \vec{q}; \vec{q}) \delta \vec{S}(\vec{k} + \vec{q}, t) - W^\nu(\vec{k}; \vec{q}) \delta \vec{S}(\vec{k}, t)] , \quad (37)$$

$$J_\nu^{(2)}[f, \delta \vec{S}] = 2 \sum_{\vec{q}} W^\nu(\vec{k} + \vec{q}; \vec{q}) \vec{g}(\vec{k}, \vec{k} + \vec{q}) \times [\vec{g}(\vec{k}, \vec{k} + \vec{q}) \times \delta \vec{S}(\vec{k} + \vec{q}, t)] , \quad (38)$$

where, for concise notation, we introduced a spin-flip vector

$$\vec{g}(\vec{k}, \vec{k}') = \begin{pmatrix} \text{Im} I_{+-}(\vec{k}, \vec{k}') \\ \text{Re} I_{+-}(\vec{k}, \vec{k}') \\ 0 \end{pmatrix} , \quad (39)$$

with  $I_{+-}(\vec{k}, \vec{k}')$  the off-diagonal element of the overlap matrix (6). This is a result of the Born approximation and the linearization with respect to the spin polarization. In general, the structure of the collision integrals depends on



the scattering process. Here, however, the scattering process enters only through  $W^\nu(\vec{k}; \vec{q})$ , the probabilities for a transition between momentum state  $\vec{k} - \vec{q}$  and  $\vec{k}$ . For electron-ionized-impurity scattering, for instance,

$$W^{ei}(\vec{k}; \vec{q}) = 2\pi N_i |U(q)|^2 \delta(\varepsilon(\vec{k} - \vec{q}) - \varepsilon(\vec{k})) , \quad (40)$$

while for electron-electron scattering,

$$\begin{aligned} W^{ee}(\vec{k}; \vec{q}) = & 4\pi |V(q)|^2 \sum_{\vec{k}'} \left\{ [1 - f(\vec{k} - \vec{q}) - f(\vec{k}' + \vec{q})] f(\vec{k}') \right. \\ & \left. + f(\vec{k} - \vec{q}) f(\vec{k}' + \vec{q}) \right\} \delta(\varepsilon(\vec{k}) + \varepsilon(\vec{k}') - \varepsilon(\vec{k} - \vec{q}) - \varepsilon(\vec{k}' + \vec{q})) , \end{aligned} \quad (41)$$

with  $U(q)$  and  $V(q)$  statically screened Coulomb potentials. Similar expressions hold for electron-phonon scattering. For electron impurity scattering, which is elastic,  $W^{ei}(\vec{k} + \vec{q}; \vec{q}) = W^{ei}(\vec{k}; \vec{q})$ ; moreover  $W^{ei}(\vec{k}; \vec{q})$  is independent of the equilibrium distribution of the spin-up and the spin-down electrons. In general, however, the transition probabilities depend on the equilibrium distribution of the electrons, and, in the case of electron-phonon scattering, also on the equilibrium distribution of the phonons.

### C. Diffusion approximation

The simple form of the collision integrals (37) and (38) suggests to conceptualize the dynamics of the non-equilibrium spin polarization in terms of spin-polarized “test” electrons, scattering off an equilibrated bath of “field” particles (impurities, electrons, and phonons). Usually this picture can be only applied to electron-impurity and electron-phonon scattering, where the scattering partners belong to different species, and not to electron-electron scattering, where the scattering partners belong to the same species. It is only within the linearized spin dynamics, which essentially treats the electrons comprising the non-equilibrium spin polarization as a separate species, that the “test-field-particle concept” can be applied to electron-electron scattering as well. We now take full advantage of the simplicity of the collision integrals and expand the collision integrals with respect to the momentum transfer  $\vec{q}$ . As a result the integro-differential equation (36) becomes a differential equation.

The on-shell spin-conserving process due to elastic electron-impurity scattering yields

$$J_B^{(0),\text{on}}[f, \delta\vec{S}] = \sum_n \sum_{i_1, \dots, i_n} C_{i_1, \dots, i_n}^{\text{ei}}(\vec{k}) \frac{\partial^n}{\partial k_{i_1} \dots \partial k_{i_n}} \delta\vec{S}(\vec{k}, t) , \quad (42)$$

whereas the inelastic spin-conserving processes due to electron-electron or electron-phonon scattering give rise to an off-shell contribution

$$J_B^{(0),\text{off}}[f, \delta\vec{S}] = \sum_{\nu=\text{ee, ep}} \sum_n \sum_{i_1, \dots, i_n} \frac{\partial^n}{\partial k_{i_1} \dots \partial k_{i_n}} C_{i_1, \dots, i_n}^\nu(\vec{k}) \delta\vec{S}(\vec{k}, t) , \quad (43)$$

where, in both cases, the moments are defined by ( $\nu = \text{ei, ee, ep}$ )

$$C_{i_1, \dots, i_n}^\nu(\vec{k}) = \frac{1}{n!} \sum_{\vec{q}} q_{i_1} \dots q_{i_n} W^\nu(\vec{k}; \vec{q}) . \quad (44)$$

The transition probability  $W^\nu(\vec{k}; \vec{q})$  depends on the precise modeling of the elementary scattering process and also on the dimensionality of the system. In the Appendix we give explicit expressions for electron-electron and electron-impurity scattering in a quantum well. Note, for inelastic scattering the differential operators act on the moments  $C_{i_1, \dots, i_n}^\nu(\vec{k})$  whereas for elastic scattering the moments are in front of the differential operators.

Equations (42) and (43) involve partial differential operators of arbitrary order. To obtain tractable equations, the expansion is in many cases truncated after the second order term (diffusion approximation). As a result, scattering processes with small momentum transfer are treated exactly whereas scattering processes with large momentum transfer are treated approximately. Because the transition probability for the (unscreened) Coulomb potential diverges for small momentum transfer, soft Coulomb scattering events dominate, and the diffusion approximation is expected to describe Coulomb scattering reasonably well. A similar reasoning applies also to electron-LO-phonon scattering. The singular behavior of the collision integrals is stronger in three than in two dimensions. The diffusion approximation

is therefore somewhat better justified for bulk than for quantum well situations. [38, 46, 48, 62] Nevertheless, our numerical results suggest that even for a quantum well the diffusion approximation gives reasonable quantitative results for the spin relaxation time.

Keeping therefore only the second order terms, we write

$$\begin{aligned}
J_B^{(0)}[f, \delta \vec{S}] &= J_B^{(0),\text{on}}[f, \delta \vec{S}] + J_B^{(0),\text{off}}[f, \delta \vec{S}] \\
&= \left[ \sum_i A^{\text{ei}}(\vec{k}) \frac{\partial}{\partial k_i} + \sum_{ij} B_{ij}^{\text{ei}}(\vec{k}) \frac{\partial^2}{\partial k_i \partial k_j} \right] \delta \vec{S}(\vec{k}, t) \\
&+ \sum_{\nu=\text{ee, ep}} \left[ \sum_i \frac{\partial}{\partial k_i} A_i^\nu(\vec{k}) + \sum_{ij} \frac{\partial^2}{\partial k_i \partial k_j} B_{ij}^\nu(\vec{k}) \right] \delta \vec{S}(\vec{k}, t) \\
&= \mathcal{D}(\vec{k}) \delta \vec{S}(\vec{k}, t) ,
\end{aligned} \tag{45}$$

where the first two terms on the rhs come from elastic scattering processes and the last two terms encode inelastic scattering events. In Eq. (45) we introduced for the first and second moments ( $\nu = \text{ei, ee, ep}$ ),

$$A_i^\nu(\vec{k}) = \sum_{\vec{q}} q_i W^\nu(\vec{k}; \vec{q}) , \tag{46}$$

$$B_{ij}^\nu(\vec{k}) = \frac{1}{2} \sum_{\vec{q}} q_i q_j W^\nu(\vec{k}; \vec{q}) , \tag{47}$$

which have the meaning of  $\vec{k}$ -dependent dynamical friction and diffusion coefficients, respectively. Within the diffusion approximation the spin-conserving (Born) collision integrals are therefore represented by a Fokker-Planck differential operator (45). Each scattering process gives rise to a particular Fokker-Planck operator, with particular dynamical friction and diffusion coefficients.

In the same spirit, expanding the spin-flip collision integral  $J_B^{(2)}[f, \delta \vec{S}]$  up to second order in the momentum transfer  $\vec{q}$ , and using  $\vec{g}(\vec{k}, \vec{k}) = 0$ , gives

$$J_B^{(2)}[f, \delta \vec{S}] = -\mathbf{R}(\vec{k}) \delta \vec{S}(\vec{k}, t) , \tag{48}$$

with a spin-flip tensor

$$\mathbf{R}(\vec{k}) = 4 \sum_{\nu=\text{ei, ee, ep}} \sum_{ij} B_{ij}^\nu(\vec{k}) \mathbf{G}_{ij}(\vec{k}) , \tag{49}$$

given in terms of the total diffusion coefficient and a tensor  $\mathbf{G}_{ij}(\vec{k})$  which describes the rate of change of the spin-flip vector  $\vec{g}(\vec{k}, \vec{k}')$ :

$$\mathbf{G}_{ij}(\vec{k}) = \begin{pmatrix} O_{ij}^{\text{yy}} & -O_{ij}^{\text{xy}} & 0 \\ -O_{ij}^{\text{yx}} & O_{ij}^{\text{xx}} & 0 \\ 0 & 0 & O_{ij}^{\text{xx}} + O_{ij}^{\text{yy}} \end{pmatrix} , \tag{50}$$

with

$$O_{ij}^{\nu\mu} = \left[ \frac{\partial}{\partial k_i} g_{\vec{k}\vec{k}'}^\nu \right]_{\vec{k}'=\vec{k}} \left[ \frac{\partial}{\partial k_j} g_{\vec{k}\vec{k}'}^\mu \right]_{\vec{k}'=\vec{k}} . \tag{51}$$

In Fig 2 we illustrate the physical content of the diffusion approximation encoded in Eqs. (45) and (48): the small “test” spin polarization  $\delta \vec{S}(\vec{k}, t)$  scatters off equilibrated “field” particles, which, depending on the scattering process, are either electrons, phonons, or impurities. Spin conserving scattering can be elastic and inelastic, because the “field” particles can absorb/emit energy, the bath has “internal degrees”. Spin non-conserving scattering, on the other hand, turns out to be elastic within the diffusion approximation.

We now introduce scaled atomic units and measure energy in units of a scaled atomic Rydberg  $\tilde{R}_0 = R_0/s$  and length in units of a scaled atomic Bohr radius  $\tilde{a}_0 = \sqrt{s}a_0$ , with  $\tilde{R}_0 \tilde{a}_0^2 = \hbar^2/2m_0$  and  $e^2 = 2\sqrt{s}\tilde{R}_0 \tilde{a}_0$ , where  $m_0$  is the bare electron mass and  $s$  is a scale factor chosen to yield  $\tilde{R}_0 = 1$  meV. Symmetry-adapted coordinates are then a

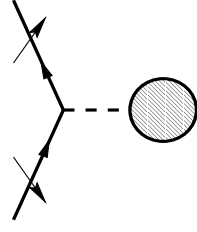


FIG. 2: Graphical illustration of the collision terms within the diffusion approximation: A “test” spin polarization scatters off a generalized bath of equilibrated “field” particles (electrons, impurities, and/or phonons). Whereas spin-flip scattering is elastic (on-shell) within our approximation, spin-conserving scattering can be on- or off-shell, i.e. the “test” spin polarization can lose/gain energy, because the “internal degrees” of the bath can absorb/emit energy.

radial coordinate  $\varepsilon = k^2$  and a generalized angle variable  $\omega$ , which, for bulk semiconductors comprises two angles, the polar angle  $\theta$  and the azimuth angle  $\phi$ , and for quantum wells is simply the polar angle  $\phi$ . Before we express the Fokker-Planck equation in these symmetry adopted coordinates, we recall that the experimentally measured quantity is the macroscopic ( $\vec{k}$ -averaged) spin polarization. Normalizing the macroscopic spin polarization to  $N_s$ , the (small) number of initially spin-polarized electrons and writing the  $\vec{k}$ -integral in symmetry-adapted coordinates, we define a “macroscopic” spin polarization (per spin polarized electron) ,

$$\begin{aligned} \delta\vec{S}(t) &= \frac{1}{N_s} \sum_{\vec{k}} \delta\vec{S}(\vec{k}, t) \\ &= \frac{1}{(2\pi)^d n_s} \int_0^\infty d\varepsilon \int d\omega J(\varepsilon) \delta\vec{S}(\varepsilon, \omega, t) , \end{aligned} \quad (52)$$

with  $d$  the dimension,  $n_s = N_s/L^d$  the density of initially spin-polarized electrons and  $J(\varepsilon)$  the energy dependent part of the Jacobian, which arises from the transformation to the symmetry-adapted coordinates. Note that due to the normalization,  $\delta\vec{S}(0)$  is a unit vector in the direction of the initial spin polarization. For bulk,  $d = 3$ ,  $d\omega = d\phi d\theta \sin\theta$ , and  $J(\varepsilon) = \sqrt{\varepsilon}/2$ , while for quantum wells  $d = 2$ ,  $d\omega = d\phi$ , and  $J(\varepsilon) = 1/2$ . Instead of setting up the Fokker-Planck equation for  $\delta\vec{S}(\varepsilon, \omega, t)$  it is more convenient to directly construct the Fokker-Planck equation for

$$\delta\vec{S}'(\varepsilon, \omega, t) = \frac{J(\varepsilon)}{(2\pi)^d n_s} \delta\vec{S}(\varepsilon, \omega, t) . \quad (53)$$

The differential operator describing spin conserving scattering processes in the Fokker-Planck equation for  $\delta\vec{S}'$  reads in symmetry-adapted coordinates

$$\begin{aligned} \mathcal{D}(\vec{k}) &= -\frac{\partial}{\partial\varepsilon} \frac{\varepsilon}{\tau_f(\varepsilon)} + \frac{\partial^2}{\partial\varepsilon^2} \frac{\varepsilon^2}{\tau_d(\varepsilon)} - \frac{1}{4\tau_\perp(\varepsilon)} \mathcal{L}^2(\omega) \\ &= \mathcal{D}(\varepsilon) - \frac{1}{4\tau_\perp(\varepsilon)} \mathcal{L}^2(\omega) , \end{aligned} \quad (54)$$

where the operator  $\mathcal{L}^2(\omega)$  denotes the total angular momentum operator in momentum space. To obtain this generic form for both bulk and quantum wells it is essential to include  $J(\varepsilon)$  into the definition of the spin polarization. The off-shell term  $\mathcal{D}(\varepsilon)$  originates from inelastic scattering events, e.g., due to electron-electron or electron-phonon scattering. The relaxation rates  $1/\tau_f(\varepsilon)$  and  $1/\tau_d(\varepsilon)$  denote the rate with which the “test” spin polarization loses energy and the rate with which the “test” spin polarization diffuses in energy space, respectively. The on-shell term, describing randomization of the angle variable, is given by the last term on the rhs of Eq. (54). It is proportional to the total on-shell relaxation rate  $1/\tau_\perp(\varepsilon)$  due to both elastic and inelastic scattering processes.

The rates characterizing the differential operator  $\mathcal{D}(\vec{k})$  are obtained from a direct calculation of the coefficients  $A_i^{\nu}(\vec{k})$  and  $B_{ij}^{\nu}(\vec{k})$  and casting the resulting differential operator  $\mathcal{D}(\vec{k})$  in the specific form given in Eq. (54). An explicit calculation of the symmetry-adapted form of the relaxation tensor  $\mathbf{R}(\vec{k})$ , which is defined in terms of the total diffusion coefficient  $B_{ij}(\vec{k})$ , due to both elastic and inelastic scattering processes, shows moreover that it can be expressed in terms of the same scattering rates. Thus, the three scattering rates  $1/\tau_f(\varepsilon)$ ,  $1/\tau_d(\varepsilon)$ , and  $1/\tau_\perp(\varepsilon)$  completely specify the two collision integrals  $J_B^{(0)}[f, \delta\vec{S}]$  and  $J_B^{(2)}[f, \delta\vec{S}]$ . In the Appendix we give explicit expressions for the relaxation rates due to electron-electron and electron-impurity scattering in a quantum well.

The dimensionless, symmetry-adapted Fokker-Planck equation for  $\delta\vec{S}'$ , which is the basis for the calculation of the spin relaxation rates presented in the next subsection, can be therefore written as

$$\begin{aligned} \frac{\partial}{\partial t} \delta\vec{S}'(\varepsilon, \omega, t) = & [\mathcal{D}(\varepsilon) - \frac{1}{4\tau_{\perp}(\varepsilon)} \mathcal{L}^2(\omega) + i\vec{\Omega}_C \cdot \vec{\mathcal{L}}(\omega)] \delta\vec{S}'(\varepsilon, \omega, t) + \vec{\Omega}_L \times \delta\vec{S}'(\varepsilon, \omega, t) \\ & + [\vec{\Omega}_{IA}(\varepsilon, \omega) + \vec{\Omega}_g(\varepsilon, \omega)] \times \delta\vec{S}'(\varepsilon, \omega, t) - \mathbf{R}(\varepsilon, \omega) \delta\vec{S}'(\varepsilon, \omega, t) . \end{aligned} \quad (55)$$

It contains spin relaxation due to motional-narrowing (DP and VG processes) and spin-flip scattering (EY process). The former arises from the combined action of the off- and on-shell spin-conserving scattering events encoded in the differential operators  $\mathcal{D}(\varepsilon)$  and  $(1/\tau_{\perp}(\omega))\mathcal{L}^2(\omega)$ , respectively, and the torque forces due to  $\vec{\Omega}_{IA}(\varepsilon, \omega)$  and  $\vec{\Omega}_g(\varepsilon, \omega)$ , while the latter originates from the spin-flip tensor  $\mathbf{R}(\varepsilon, \omega)$ . Note that if more than one scattering process is considered, the relaxation rates characterizing the differential operators and the spin-flip tensor are total relaxation rates, due to whatever elastic and inelastic scattering processes are included in the model. The orbital motion of the electrons, which leads to a quenching of the motional-narrowing spin relaxation processes, is given by the term  $i\vec{\Omega}_C \cdot \vec{\mathcal{L}}(\omega)$  on the rhs of Eq. (55). In the next subsection we develop a scheme which separates the fast spin conserving scattering processes from the slow spin decay causing processes and directly yields the time evolution of the macroscopic spin polarization.

#### D. Multiple time scale analysis

The Fokker-Planck equation (55) determines the time evolution of the non-equilibrium spin polarization on the fast, spin-conserving time scale, where randomization of the angle variables (direction of the momentum) and energy relaxation and diffusion occurs, and on the long time scale, where spin non-conserving processes lead to the decay of the spin polarization. The two time scales are well separated. The fast, spin conserving stage, whose scale is given by the first term on the rhs of Eq. (55) and therefore by the off- and on-shell relaxation times (as well as the time it takes to complete one cyclotron orbit), terminates in a quasi-stationary state, which then evolves on the time scales set by the Elliott-Yafet term, the torque forces due to the spin off-diagonal Hamiltonian matrix elements, and the external magnetic field (Larmor precession). Experimentally relevant is usually the time evolution on the long time scale. In this subsection we employ therefore a multiple time scale approach to extract from the Fokker-Planck equation (55) a Bloch equation, which controls the time evolution of the macroscopic ( $\vec{k}$ -averaged) spin polarization on the long time scale. To simplify the notation, we suppress the prime. It is understood that the spin polarization  $\delta\vec{S}(\varepsilon, \omega, t)$  contains the factor  $J(\varepsilon)/(2\pi)^d n_s$ .

As a preparatory step we first consider that part of the Fokker-Planck equation (55), which is spin-conserving:

$$\frac{\partial}{\partial t} \delta\vec{S}(\varepsilon, \omega, t) = [\mathcal{D}(\varepsilon) - \frac{1}{4\tau_{\perp}(\varepsilon)} \mathcal{L}^2(\omega) + i\vec{\Omega}_C \cdot \vec{\mathcal{L}}(\omega)] \delta\vec{S}(\varepsilon, \omega, t) . \quad (56)$$

To find the stationary solution of Eq. (56) we set the lhs to zero, write

$$\delta\vec{S}_{st}(\varepsilon, \omega) = p(\varepsilon) \vec{\Psi}_{st}(\varepsilon, \omega) , \quad (57)$$

with  $p(\varepsilon)$  defined by

$$\mathcal{D}(\varepsilon) p(\varepsilon) = 0 , \quad (58)$$

and obtain for the auxiliary vector  $\vec{\Psi}_{st}(\varepsilon, \omega)$  the differential equation

$$[\mathcal{D}^*(\varepsilon) - \frac{1}{4\tau_{\perp}(\varepsilon)} \mathcal{L}^2(\omega) + i\vec{\Omega}_C \cdot \vec{\mathcal{L}}(\omega)] \vec{\Psi}_{st}(\varepsilon, \omega) = 0 , \quad (59)$$

with

$$\mathcal{D}^*(\varepsilon) = \frac{\varepsilon}{\tau_f(\varepsilon)} \frac{\partial}{\partial \varepsilon} + \frac{\varepsilon^2}{\tau_d(\varepsilon)} \frac{\partial^2}{\partial \varepsilon^2} , \quad (60)$$

the adjoint operator to  $\mathcal{D}(\varepsilon)$ . The simplest solution of (59) is a constant vector

$$\vec{\Psi}_{st}(\varepsilon, \omega) = \vec{e} , \quad (61)$$

giving rise to a stationary solution

$$\delta\vec{S}_{st}(\varepsilon, \omega) = p(\varepsilon)\vec{e} . \quad (62)$$

The particular form of  $p(\varepsilon)$  does not matter at this point.

We now turn to the full Fokker-Planck equation (55). The spin-conserving stage of the time evolution, described by the first term on the rhs, occurs on a very fast time scale and is usually experimentally not resolved. Hence, it is not necessary to explicitly keep track of it. Instead, it is sufficient to use the final state of the fast, spin-conserving time evolution, i.e. the stationary solution of the spin conserving part of the Fokker-Planck equation (viz: Eqs. (56)–(62)), as an initial state for the time evolution on the slow time scale, where spin decay occurs. We write the initial condition therefore as

$$\delta\vec{S}_{st}(\varepsilon, \omega, 0) = p(\varepsilon)\vec{e} , \quad (63)$$

where  $\vec{e}$  is now the direction of the initial spin polarization. This initial condition is general enough, because neither electrical nor optical spin injection produce anisotropic initial spin polarizations. Accordingly, we write for arbitrary times

$$\delta\vec{S}(\varepsilon, \omega, t) = p(\varepsilon)\vec{\Psi}(\varepsilon, \omega, t) , \quad (64)$$

where the time-dependent auxiliary vector  $\vec{\Psi}(\varepsilon, \omega, t)$  satisfies now the time-dependent equation

$$\begin{aligned} \frac{\partial}{\partial t}\vec{\Psi}(\varepsilon, \omega, t) &= [\mathcal{D}^*(\varepsilon) - \frac{1}{4\tau_{\perp}(\varepsilon)}\mathcal{L}^2(\omega) + i\vec{\Omega}_C \cdot \vec{\mathcal{L}}(\omega)]\vec{\Psi}(\varepsilon, \omega, t) \\ &+ \left( \vec{\Omega}_L + \vec{\Omega}_{IA}(\varepsilon, \omega) + \vec{\Omega}_g(\varepsilon, \omega) \right) \times \vec{\Psi}(\varepsilon, \omega, t) - \mathbf{R}(\varepsilon, \omega)\vec{\Psi}(\varepsilon, \omega, t) , \end{aligned} \quad (65)$$

with an initial condition  $\vec{\Psi}(\varepsilon, \omega, 0) = \vec{e}$ . Note that Eq. (65) is the adjoint Fokker-Planck equation. The function  $p(\varepsilon)$  satisfies the homogeneous differential equation (58) and is therefore defined only up to a normalization constant. From the initial condition for the macroscopic spin polarization,  $\delta\vec{S}(0) = \vec{e}$ , we conclude that  $p(\varepsilon)$  has to be normalized according to  $\int d\varepsilon \int d\omega p(\varepsilon) = 1$  (recall that we redefined  $\delta\vec{S}$  such that it contains the factor  $J(\varepsilon)/(2\pi)^d n_s$ ). Thus,  $d\varepsilon \int d\omega p(\varepsilon)$  can be interpreted as the probability density for finding a spin-polarized “test” electron in the energy interval  $[\varepsilon, \varepsilon + d\varepsilon]$ .

To proceed further we scale each term in Eq. (65) to its typical value. In the case of degenerate electrons the typical values would be the ones at the Fermi energy, whereas for non-degenerate electrons the typical values could be the ones at the average thermal energy. Denoting typical values by a caret, we introduce scaled quantities  $t' = t/\hat{t}$ ,  $\tau'_f = \tau_f/\hat{\tau}_f$ ,  $\tau'_d = \tau_d/\hat{\tau}_d$ ,  $\tau'_\perp = \tau_\perp/\hat{\tau}_\perp$ ,  $\vec{\Omega}'_{IA} = \vec{\Omega}_{IA}\hat{\tau}_{IA}$ ,  $\vec{\Omega}'_g = \vec{\Omega}_g\hat{\tau}_g$ ,  $\vec{\Omega}'_L = \vec{\Omega}_L\hat{\tau}_L$ ,  $\mathbf{R}' = \mathbf{R}\hat{\tau}_R$ , and  $\vec{\Omega}'_C = \vec{\Omega}_C\hat{\tau}_C$ . The rescaled equation for  $\vec{\Psi}'(\varepsilon, \omega, t')$  becomes (suppressing the arguments of the various functions)

$$\begin{aligned} \frac{\partial}{\partial t'}\vec{\Psi}' &= \left[ \frac{\hat{t}}{\hat{\tau}_f} \frac{\varepsilon}{\tau'_f} \frac{\partial}{\partial \varepsilon} + \frac{\hat{t}}{\hat{\tau}_d} \frac{\varepsilon^2}{\tau'_d} \frac{\partial^2}{\partial \varepsilon^2} - \frac{\hat{t}}{\hat{\tau}_\perp} \frac{1}{4\tau'_\perp} \mathcal{L}^2 + \frac{\hat{t}}{\hat{\tau}_C} i\vec{\Omega}'_C \cdot \vec{\mathcal{L}} \right] \vec{\Psi}' \\ &+ \frac{\hat{t}}{\hat{\tau}_L} \vec{\Omega}'_L \times \vec{\Psi}' + \frac{\hat{t}}{\hat{\tau}_{IA}} \vec{\Omega}'_{IA} \times \vec{\Psi}' + \frac{\hat{t}}{\hat{\tau}_g} \vec{\Omega}'_g \times \vec{\Psi}' - \frac{\hat{t}}{\hat{\tau}_R} \mathbf{R}' \vec{\Psi}' . \end{aligned} \quad (66)$$

We identify three time scales. A fast time scale given by the spin conserving relaxation times  $\hat{\tau}_i$  ( $i = f, d, \perp$ ) and the time it takes to complete a cyclotron orbit  $\hat{\tau}_C$ , an intermediate time scale given by the time it takes to complete a precession around the intrinsic magnetic fields (due to the spin off-diagonal Hamiltonian matrix elements)  $\hat{\tau}_{IA}$  and  $\hat{\tau}_g$ , and a long time scale, on which Larmor precession and spin-flip scattering occur,  $\hat{\tau}_L$  and  $\hat{\tau}_R$ , respectively. For representative experimental set-ups, the typical time scale  $\hat{t}$ , on which the spin polarization has to be tracked (“observation time”), and the three typical intrinsic time scales obey the following ordering:  $\frac{\hat{t}}{\hat{\tau}_f}, \frac{\hat{t}}{\hat{\tau}_d}, \frac{\hat{t}}{\hat{\tau}_\perp}, \frac{\hat{t}}{\hat{\tau}_C} = O(\eta^{-1})$ ,  $\frac{\hat{t}}{\hat{\tau}_{IA}}, \frac{\hat{t}}{\hat{\tau}_g} = O(\eta^0)$ , and  $\frac{\hat{t}}{\hat{\tau}_L}, \frac{\hat{t}}{\hat{\tau}_R} = O(\eta^1)$  where we introduced a small parameter  $\eta$ . Accordingly, we classify each term in Eq. (66) by the smallness parameter  $\eta$ . Suppressing the primes, Eq. (66) is rewritten as

$$\frac{\partial}{\partial t}\vec{\Psi} = \frac{1}{\eta} [\mathcal{D}^* - \frac{1}{4\tau_\perp} \mathcal{L}^2 + i\vec{\Omega}_C \cdot \vec{\mathcal{L}}] \vec{\Psi} + (\vec{\Omega}_{IA} + \vec{\Omega}_g) \times \vec{\Psi} + \eta \vec{\Omega}_L \times \vec{\Psi} - \eta \mathbf{R} \vec{\Psi} . \quad (67)$$

Equation (67) is in a form where fast and slow processes can be clearly identified. The fast spin-conserving terms and the orbital motion enter in order  $\eta^{-1}$ , the precession around the internal magnetic fields enters in order  $\eta^0$ , whereas

the Larmor precession and the spin-flip scattering terms appear in order  $\eta^1$ . Naturally, taking as much advantage as possible of the existence of the small parameter  $\eta$ , the first thought is to expand all quantities with respect to  $\eta$  and apply perturbation theory. The structure of Eq. (67) indicates however that regular perturbation theory will lead to non-uniformity in the long-time regime, i.e. precisely in that regime, which we are interested in. To obtain the correct long-time behavior of the solution of Eq. (67) a multiple time scale approach is required.

In the spirit of multiple time scale perturbation theory [63], we consider therefore  $\vec{\Psi}$  as a function of three time variables  $t_n = \eta^n t$ ,  $n = -1, 0, 1$ , which are assumed to be independent, and substitute a second order expansion of the form

$$\vec{\Psi}(\varepsilon, \omega, t) = \vec{\Psi}^{(0)}(\varepsilon, \omega, t_{-1}, t_0, t_1) + \eta \vec{\Psi}^{(1)}(\varepsilon, \omega, t_{-1}, t_0, t_1) + \eta^2 \vec{\Psi}^{(2)}(\varepsilon, \omega, t_{-1}, t_0, t_1) \quad (68)$$

into Eq. (67), where the time derivative is extended to  $\partial_t = \eta^{-1} \partial_{t_{-1}} + \partial_{t_0} + \eta \partial_{t_1}$ . Equating coefficients of like powers of  $\eta$  yields an hierarchy of equations for the functions  $\vec{\Psi}^{(n)}$ . Up to  $O(\eta)$  they read

$$\frac{\partial}{\partial t_{-1}} \vec{\Psi}^{(0)} = [\mathcal{D}^* - \frac{1}{4\tau_{\perp}} \mathcal{L}^2 + i\vec{\Omega}_C \cdot \vec{\mathcal{L}}] \vec{\Psi}^{(0)}, \quad (69)$$

$$\frac{\partial}{\partial t_{-1}} \vec{\Psi}^{(1)} + \frac{\partial}{\partial t_0} \vec{\Psi}^{(0)} = [\mathcal{D}^* - \frac{1}{4\tau_{\perp}} \mathcal{L}^2 + i\vec{\Omega}_C \cdot \vec{\mathcal{L}}] \vec{\Psi}^{(1)} + (\vec{\Omega}_{IA} + \vec{\Omega}_g) \times \vec{\Psi}^{(0)}, \quad (70)$$

$$\begin{aligned} \frac{\partial}{\partial t_{-1}} \vec{\Psi}^{(2)} + \frac{\partial}{\partial t_0} \vec{\Psi}^{(1)} + \frac{\partial}{\partial t_1} \vec{\Psi}^{(0)} &= [\mathcal{D}^* - \frac{1}{4\tau_{\perp}} \mathcal{L}^2 + i\vec{\Omega}_C \cdot \vec{\mathcal{L}}] \vec{\Psi}^{(2)} + (\vec{\Omega}_{IA} + \vec{\Omega}_g) \times \vec{\Psi}^{(1)} \\ &+ \vec{\Omega}_L \times \vec{\Psi}^{(0)} - \mathbf{R} \vec{\Psi}^{(0)}. \end{aligned} \quad (71)$$

For the analysis of this set of equations it is convenient to split  $\vec{\Psi}^{(n)}$  into an angle averaged and a remaining part,

$$\vec{\Psi}^{(n)}(\varepsilon, \omega, t_{-1}, t_0, t_1) = \vec{a}^{(n)}(\varepsilon, t_{-1}, t_0, t_1) + \delta \vec{a}^{(n)}(\varepsilon, \omega, t_{-1}, t_0, t_1), \quad (72)$$

where  $\vec{a}^{(n)} = \langle \vec{\Psi}^{(n)} \rangle_{\omega}$ , with an angle average defined by

$$\langle (\dots) \rangle_{\omega} = \int d\omega (\dots) \quad (73)$$

and  $\langle \delta \vec{a}^{(n)} \rangle_{\omega} = 0$  by definition. Since the angle variables are periodic this partitioning is always possible. From the initial condition,  $\vec{\Psi}(\varepsilon, \omega, 0) = \vec{e}$ , we infer the initial conditions  $\vec{a}^{(n)}(\varepsilon, 0, 0, 0) = \vec{e} \delta_{n,0}$ , and  $\delta \vec{a}^{(n)}(\varepsilon, \omega, 0, 0, 0) = 0$ . Recalling that the factor  $J(\varepsilon)/(2\pi)^d n_s$  is included in the definition of  $\delta \vec{S}(\varepsilon, \omega, t)$ , the macroscopic non-equilibrium spin polarization defined in Eq. (52) can now be rewritten as

$$\delta \vec{S}(t) = \int_0^{\infty} d\varepsilon p(\varepsilon) \vec{a}(\varepsilon, t) = \langle \vec{a}(\varepsilon, t) \rangle_{\varepsilon}, \quad (74)$$

where we defined an energy average

$$\langle (\dots) \rangle_{\varepsilon} = \int_0^{\infty} d\varepsilon p(\varepsilon) (\dots). \quad (75)$$

Note that the function  $p(\varepsilon)$ , which determines the terminating state of the fast, spin-conserving time evolution, enters here naturally as a weight function. Formally, the weight function appears in our theory because of the Ansatz (64), which enabled us to switch to the adjoint Fokker-Planck equation. The expansion of  $\vec{\Psi}$  implies an analogous expansion for the macroscopic ( $\vec{k}$ -averaged) spin polarization:

$$\delta \vec{S}(t) = \delta \vec{S}^{(0)}(t_{-1}, t_0, t_1) + \eta \delta \vec{S}^{(1)}(t_{-1}, t_0, t_1) + \eta^2 \delta \vec{S}^{(2)}(t_{-1}, t_0, t_1). \quad (76)$$

We now calculate the leading order contribution  $\delta \vec{S}^{(0)}(t)$  uniformly valid for all times. As a result, we will obtain a Bloch equation which determines the long time behavior of the macroscopic spin polarization.

### 1. $O(\eta^{-1})$ equation

With the substitution (72), the  $O(\eta^{-1})$  equation (69) splits into two independent equations, one for the angle averaged part and one for the angle dependent part.

$$\frac{\partial}{\partial t_{-1}} \bar{a}^{(0)} = \mathcal{D}^* \bar{a}^{(0)} , \quad (77)$$

$$\frac{\partial}{\partial t_{-1}} \delta \bar{a}^{(0)} = [\mathcal{D}^* - \frac{1}{4\tau_{\perp}} \mathcal{L}^2] \delta \bar{a}^{(0)} . \quad (78)$$

Solutions of Eqs. (77) and (78) compatible with the two initial conditions,  $\bar{a}^{(0)}(\varepsilon, 0, 0, 0) = \bar{e}$  and  $\delta \bar{a}^{(0)}(\varepsilon, 0, 0, 0) = 0$ , are  $\bar{a}^{(0)}(\varepsilon, t_{-1}, t_0, t_1) = \bar{a}^{(0)}(t_0, t_1)$ , with  $\bar{a}^{(0)}(0, 0) = \bar{e}$ , and  $\delta \bar{a}^{(0)}(\varepsilon, \omega, t_{-1}, t_0, t_1) = 0$ . Using Eqs. (74) and (76), we find therefore

$$\delta \vec{S}^{(0)}(t_0, t_1) = \bar{a}^{(0)}(t_0, t_1) , \quad (79)$$

that is, due to our choice of the initial condition, the macroscopic zeroth order spin polarization is independent of the fast spin conserving time scale  $t_{-1}$  and solely evolves on the long time scales  $t_0$  and  $t_1$ .

### 2. $O(\eta^0)$ equation

To determine the time evolution of the macroscopic spin polarization on the long time scales  $t_0$  and  $t_1$ , we study the  $O(\eta^0)$  equation. Substituting (72) into Eq. (70), the latter splits into two independent equations:

$$\frac{\partial}{\partial t_{-1}} \bar{a}^{(1)} = -\frac{\partial}{\partial t_0} \bar{a}^{(0)} + \mathcal{D}^* \bar{a}^{(1)} , \quad (80)$$

$$\frac{\partial}{\partial t_{-1}} \delta \bar{a}^{(1)} = [\mathcal{D}^* - \frac{1}{4\tau_{\perp}} \mathcal{L}^2 + i\vec{\Omega}_C \cdot \vec{\mathcal{L}}] \delta \bar{a}^{(1)} + (\vec{\Omega}_{IA} + \vec{\Omega}_g) \times \bar{a}^{(0)} , \quad (81)$$

where we have used  $\delta \bar{a}^{(0)} = 0$ ,  $\langle \vec{\Omega}_{IA} \rangle_{\omega} = 0$ , and  $\langle \vec{\Omega}_g \rangle_{\omega} = 0$ . Applying the energy average  $\langle \langle \dots \rangle \rangle_{\varepsilon}$  on both sides of Eq. (80) yields

$$\frac{\partial}{\partial t_{-1}} \delta \vec{S}^{(1)} = -\frac{\partial}{\partial t_0} \delta \vec{S}^{(0)} - \langle \mathcal{D}^* \bar{a}^{(1)} \rangle_{\varepsilon} , \quad (82)$$

which, using in the second term partial integration and the definition of  $p(\varepsilon)$ , reduces to

$$\frac{\partial}{\partial t_{-1}} \delta \vec{S}^{(1)} = -\frac{\partial}{\partial t_0} \delta \vec{S}^{(0)} . \quad (83)$$

The vanishing of the second term on the rhs of Eq. (82) is the result of the spin conservation of the differential operator  $\mathcal{D}$ , which in turn is ensured by the identity

$$\left. \frac{d}{d\varepsilon} \frac{\varepsilon^2}{\tau_d(\varepsilon)} \right|_{\varepsilon=0} = \left. \frac{\varepsilon}{\tau_f(\varepsilon)} \right|_{\varepsilon=0} . \quad (84)$$

It is crucial to note that the rhs of Eq. (83) is independent of the fast time  $t_{-1}$ , because  $\bar{a}^{(0)}$  is independent of  $t_{-1}$ . Integrating Eq. (83) with respect to  $t_{-1}$ , the rhs therefore gives rise to a secular term, i.e. a term which is proportional to  $t_{-1}$ . As a result,  $\delta \vec{S}^{(1)}$  can be larger than  $\delta \vec{S}^{(0)}$  for sufficiently large times. The expansion (76) would be valid only for short times, i.e. the expansion is non-uniform. Within multiple time scale perturbation theory, secular terms can be avoided by an appropriate choice of the time evolutions on the various time scales. The secular term in Eq. (83) can be particularly simply removed by forcing the rhs to be zero, which gives rise to the condition

$$\int_0^{\infty} d\varepsilon p(\varepsilon) \frac{\partial}{\partial t_0} \bar{a}^{(0)}(t_0, t_1) = 0 . \quad (85)$$

That is,  $\bar{a}^{(0)}(t_0, t_1) = \bar{a}^{(0)}(t_1)$ , which, using Eq. (79), leads to  $\delta \vec{S}^{(0)}(t_1) = \bar{a}^{(0)}(t_1)$ , i.e. the time evolution of the zeroth order macroscopic spin polarization (and therefore the spin decay) occurs solely on the long time scale  $t_1$ . Since the rhs of Eq. (83) is made to vanish, we also obtain  $\delta \vec{S}^{(1)}(t_{-1}, t_0, t_1) = \delta \vec{S}^{(1)}(t_0, t_1)$ , i.e.,  $\delta \vec{S}^{(1)}$  is independent of the fast time variable  $t_{-1}$ . Using the definition of  $\delta \vec{S}^{(1)}$ , we furthermore conclude that  $\bar{a}^{(1)}(\varepsilon, t_{-1}, t_0, t_1) = \bar{a}^{(1)}(\varepsilon, t_0, t_1)$ . Both results we need in the analysis of the  $O(\eta^1)$  equations, which is necessary to determine the time evolution on the remaining time scale  $t_1$ .

### 3. $O(\eta^1)$ equations

To investigate the  $O(\eta^1)$  equation (71), we substitute (72) into (71). Averaging over the angle and energy, we find

$$\frac{\partial}{\partial t_{-1}} \delta \vec{S}^{(2)} = -\frac{\partial}{\partial t_0} \delta \vec{S}^{(1)} - \frac{\partial}{\partial t_1} \delta \vec{S}^{(0)} + \langle (\vec{\Omega}_{IA} + \vec{\Omega}_g) \times \delta \vec{a}^{(1)} \rangle_{\varepsilon, \omega} + \vec{\Omega}_L \times \delta \vec{S}^{(0)} - \langle \mathbf{R} \rangle_{\varepsilon, \omega} \delta \vec{S}^{(0)} , \quad (86)$$

where we used  $\delta \vec{a}^{(0)} = 0$ ,  $\langle \vec{\Omega}_{IA} \rangle_{\varepsilon, \omega} = 0$ ,  $\langle \vec{\Omega}_g \rangle_{\varepsilon, \omega} = 0$ , and  $\langle \mathcal{D}^* \vec{a}^{(2)} \rangle_{\varepsilon, \omega} = 0$ . The third term on the rhs of (86) contains  $\delta \vec{a}^{(1)}$  which has to be obtained from Eq. (81), the angle dependent part of the  $O(\eta^0)$  equation. Before we proceed with the analysis of Eq. (86) let us therefore turn to Eq. (81). Integration of Eq. (81) with respect to the fast time  $t_{-1}$  produces a secular term,  $(\vec{\Omega}_{IA} + \vec{\Omega}_g) \times \vec{a}^{(0)} t_{-1}$ , which cannot be removed, because both  $\vec{\Omega}_{IA} + \vec{\Omega}_g$  and  $\vec{a}^{(0)}$  are finite. A way to avoid the resulting non-uniformity is to demand

$$\frac{\partial}{\partial t_{-1}} \delta \vec{a}^{(1)} = 0 , \quad (87)$$

i.e., to enforce  $\delta \vec{a}^{(1)}(\varepsilon, \omega, t_{-1}, t_0, t_1) = \delta \vec{a}^{(1)}(\varepsilon, \omega, t_0, t_1)$ , which reduces Eq. (81) to

$$[\mathcal{D}^* - \frac{1}{4\tau_{\perp}} \mathcal{L}^2 + i\vec{\Omega}_C \cdot \vec{\mathcal{L}}] \delta \vec{a}^{(1)} + (\vec{\Omega}_{IA} + \vec{\Omega}_g) \times \delta \vec{S}^{(0)} = 0 , \quad (88)$$

where we used in the last term  $\vec{a}^{(0)}(t_1) = \delta \vec{S}^{(0)}(t_1)$ . The condition (87) is reminiscent of the quasi-stationarity assumption usually invoked in the calculation of the D'yakonov-Perel' relaxation rates. [7] The multiple time scale approach enables us to identify the time scale on which this assumption holds.

We now return to Eq. (86). Since  $\delta \vec{a}^{(1)}$ ,  $\delta \vec{S}^{(1)}$ , and  $\delta \vec{S}^{(0)}$  are independent of the fast time variable  $t_{-1}$ , the whole rhs of Eq. (86) is independent of  $t_{-1}$ . Integration with respect to  $t_{-1}$  thus gives rise to a secular term which has to be removed. We force therefore the rhs of Eq. (86) to vanish which can be certainly accomplished if we separately demand

$$\frac{\partial}{\partial t_0} \delta \vec{S}^{(1)} = 0 , \quad (89)$$

$$\frac{\partial}{\partial t_1} \delta \vec{S}^{(0)} = \vec{\Omega}_L \times \delta \vec{S}^{(0)} + \langle (\vec{\Omega}_{IA} + \vec{\Omega}_g) \times \delta \vec{a}^{(1)} \rangle_{\varepsilon, \omega} - \langle \mathbf{R} \rangle_{\varepsilon, \omega} \delta \vec{S}^{(0)} . \quad (90)$$

From the first equation we find

$$\int_0^\infty d\varepsilon p(\varepsilon) \frac{\partial}{\partial t_0} \vec{a}^{(1)}(\varepsilon, t_0, t_1) = 0 , \quad (91)$$

that is  $\vec{a}^{(1)}$  is independent of  $t_0$ , a result which we need below. The second equation is already a precursor of the Bloch equation for  $\delta \vec{S}^{(0)}(t_1)$ . Although it determines  $\delta \vec{S}^{(0)}(t_1)$  for a given  $\delta \vec{a}^{(1)}(t_0, t_1)$ , it is however not yet a Bloch equation because, at this point of the calculation,  $\delta \vec{a}^{(1)}$  is still a function of  $t_1$  and  $t_0$ .

To obtain a closed Bloch equation on the time scale  $t_1$  alone, we now examine the  $t_0$  dependence of  $\delta \vec{a}^{(1)}(t_0, t_1)$ . Towards that end, we consider the angle dependent part of the  $O(\eta^1)$  equation (71), which reads

$$\begin{aligned} \frac{\partial}{\partial t_{-1}} \delta \vec{a}^{(2)} &= [\mathcal{D}^* - \frac{1}{4\tau_{\perp}} \mathcal{L}^2 + i\vec{\Omega}_C \cdot \vec{\mathcal{L}}] \delta \vec{a}^{(2)} \\ &+ (\vec{\Omega}_{IA} + \vec{\Omega}_g) \times \vec{a}^{(1)} + (\vec{\Omega}_{IA} + \vec{\Omega}_g) \times \delta \vec{a}^{(1)} - \langle (\vec{\Omega}_{IA} + \vec{\Omega}_g) \times \delta \vec{a}^{(1)} \rangle_{\omega} \\ &- \mathbf{R} \vec{a}^{(0)} + \langle \mathbf{R} \rangle_{\omega} \vec{a}^{(0)} - \frac{\partial}{\partial t_0} \delta \vec{a}^{(1)} . \end{aligned} \quad (92)$$

Except for the first term, all terms on the rhs are independent of  $t_{-1}$ , and therefore give rise to secular terms. To remove the secular terms, we set the undesired terms on the rhs to zero

$$\begin{aligned} \frac{\partial}{\partial t_0} \delta \vec{a}^{(1)} &= (\vec{\Omega}_{IA} + \vec{\Omega}_g) \times \delta \vec{a}^{(1)} - \langle (\vec{\Omega}_{IA} + \vec{\Omega}_g) \times \delta \vec{a}^{(1)} \rangle_{\omega} \\ &- [\mathbf{R} - \langle \mathbf{R} \rangle_{\omega}] \vec{a}^{(0)} + (\vec{\Omega}_{IA} + \vec{\Omega}_g) \times \vec{a}^{(1)} . \end{aligned} \quad (93)$$



The last two terms on the rhs of this equation are independent of  $t_0$  and therefore again give rise to a secular term if Eq. (93) is integrated with respect to  $t_0$ . In general, the last two terms are finite. Thus, to avoid non-uniformity we demand that  $\delta\vec{a}^{(1)}$  is independent of  $t_0$ , that is, we enforce  $\delta\vec{a}^{(1)}(\varepsilon, \omega, t_0, t_1) = \delta\vec{a}^{(1)}(\varepsilon, \omega, t_1)$ . With this constraint, Eq. (93) could be used to determine  $\vec{a}^{(1)}(\varepsilon, t_1)$  and eventually  $\delta\vec{S}^{(1)}(t_1)$ .

Because  $\delta\vec{a}^{(1)}$  is independent of  $t_0$ , Eq. (90) is in fact a Bloch equation on the time scale  $t_1$  alone. The function  $\delta\vec{a}^{(1)}(\varepsilon, \omega, t_1)$  satisfies Eq. (88), which, through  $\delta\vec{S}^{(0)}(t_1)$ , contains  $t_1$  only as a parameter. Therefore, the function  $\delta\vec{a}^{(1)}(\varepsilon, \omega, t_1)$  instantaneously adjusts to the function  $\delta\vec{S}^{(0)}(t_1)$ , which, in this sense, acts like a “slave field” for  $\delta\vec{a}^{(1)}(\varepsilon, \omega, t_1)$ .

To make the equations determining the decay of the macroscopic spin polarization explicit, we recall  $t_n = \eta^n t$  and go back to the original, unscaled time variable and functions. As a result Eq. (90) becomes a Bloch equation for the macroscopic spin polarization,

$$\frac{\partial}{\partial t} \delta\vec{S}^{(0)}(t) = \vec{\Omega}_L \times \delta\vec{S}^{(0)}(t) - [\mathbf{\Gamma}_{\text{EY}} + \mathbf{\Gamma}_{\text{MN}}] \delta\vec{S}^{(0)}(t) , \quad (94)$$

with initial condition  $\delta\vec{S}(0) = \vec{e}$ . The Elliott-Yafet and motional-narrowing spin relaxation tensors are given by

$$\mathbf{\Gamma}_{\text{EY}} = \langle \mathbf{R} \rangle_{\varepsilon, \omega}, \quad (95)$$

$$\mathbf{\Gamma}_{\text{MN}} \delta\vec{S}^{(0)}(t) = -\langle (\vec{\Omega}_{IA} + \vec{\Omega}_g) \times \delta\vec{a}^{(1)} \rangle_{\varepsilon, \omega} , \quad (96)$$

respectively, and  $\delta\vec{a}^{(1)}$  is obtained from Eq. (88), which for convenience we state here again

$$[\mathcal{D}^* - \frac{1}{4\tau_{\perp}} \mathcal{L}^2 + i\vec{\Omega}_C \cdot \vec{\mathcal{L}}] \delta\vec{a}^{(1)} + (\vec{\Omega}_{IA} + \vec{\Omega}_g) \times \delta\vec{S}^{(0)} = 0 .$$

Equations (94) – (96) are the main result of this section. They control the time evolution of the macroscopic spin polarization on the long time scale, where spin relaxation, i.e., decay, occurs. Equation (96) is an implicit definition of the spin relaxation tensor  $\mathbf{\Gamma}_{\text{MN}}$ . The explicit form of  $\mathbf{\Gamma}_{\text{MN}}$  can be obtained by inserting the solution of Eq. (88), which is always linear in  $\delta\vec{S}(t)$ , and performing the angle and energy averages. In the next section we illustrate this procedure for a doped quantum well subject to a small magnetic field.

The macroscopic spin relaxation tensor contains the EY process and the motional-narrowing (DP and VG) processes. Due to the different angle dependences of the two main motional-narrowing spin relaxation processes, DP and VG processes, Eq. (96) splits for isotropic semiconductors into two separate terms,  $\mathbf{\Gamma}_{\text{DP}}$  and  $\mathbf{\Gamma}_{\text{VG}}$ . Accordingly, for isotropic semiconductors, a Matthiessen-type rule holds for the total spin relaxation tensor,  $\mathbf{\Gamma} = \mathbf{\Gamma}_{\text{EY}} + \mathbf{\Gamma}_{\text{DP}} + \mathbf{\Gamma}_{\text{VG}}$ , and, as a consequence, for the spin relaxation rates, which are the diagonal elements of the relaxation tensors. [10] The quenching of the motional-narrowing processes due to the orbital motion is contained in Eq. (88) through the term proportional to  $\vec{\Omega}_C$ . [8, 10] The on- and off-shell relaxation rates appearing in Eq. (88) are total relaxation rates due to whatever scattering processes are included. A Matthiessen rule holds separately for the on- and off-shell rates. The energy average  $\langle (\dots) \rangle_{\varepsilon}$  is defined in Eq. (75). Most importantly, it contains a weight function  $p(\varepsilon)$  defined as the solution of Eq. (58). This function describes the energy dependence of the quasi-stationary spin polarization which appears on the short time scale because of fast, spin-conserving inelastic scattering processes, i.e. because of energy relaxation and diffusion. Once the quasi-stationary spin polarization is established, it slowly decays on the long time scale set by the spin non-conserving terms in the Fokker-Planck equation.

#### IV. APPLICATION TO QUANTUM WELL STRUCTURES

In this section we apply the formalism to a quantum well at low enough temperatures, where electron-impurity and electron-electron scattering dominate. We are here particularly interested in the effects of Pauli blocking and inelasticity. For illustration, we focus therefore only on the DP process, which, for small to moderate magnetic fields, is usually the dominant spin relaxation process. Moreover, if the magnetic field is small enough, the time  $\hat{\tau}_C$  it takes to complete a cyclotron orbit is much longer than any of the intrinsic scattering times  $\hat{\tau}_i$  ( $i = f, d, \perp$ ), and the quenching effect of the magnetic field can be ignored.

We consider a symmetric GaAs quantum well, grown in the [001] direction, which is also the quantization axis for the electron spin. Due to the assumed structural symmetry, there is only bulk inversion asymmetry giving rise to DP spin relaxation. [40] As in the bulk case, we treat the two states at the conduction band minimum explicitly and include a large set of states perturbatively, up to third order, to include the effect of bulk inversion asymmetry. For energies close to the band minimum, the Hamiltonian for the quantum well can be cast into the form (1). The

spin off-diagonal term in the quantum well Hamiltonian is the bulk spin off-diagonal term averaged over the envelope function of the conduction subband. Assuming for simplicity infinite confinement and restricting the calculation to the lowest conduction subband, we find (neglecting cubic terms in  $\vec{k}$ )

$$\begin{aligned} \hbar \vec{\Omega}_{IA}^{QW}(\varepsilon, \phi) &= 2\delta_0 \sqrt{\varepsilon} \left(\frac{\pi}{L}\right)^2 \begin{pmatrix} -\cos \phi \\ \sin \phi \\ 0 \end{pmatrix} \\ &= \frac{1}{\tau_{IA}(\varepsilon)} \vec{\kappa}_{IA}(\phi) , \end{aligned} \quad (97)$$

where we have defined a precession rate  $1/\tau_{IA}(\varepsilon) = C_{IA}^{QW} \sqrt{\varepsilon}$  with  $C_{IA}^{QW} = 2\delta_0(\pi/L)^2$ .

Since we are only interested in the DP spin relaxation tensor, we neglect in Eq. (88) the torque force due to  $\vec{\Omega}_g$ . Because we furthermore assume small magnetic fields, we also ignore the orbital motion of the electrons. The separation Ansatz,

$$\delta \vec{a}^{(1)}(\varepsilon, \phi, t) = \tau(\varepsilon) \vec{\kappa}_{IA}(\phi) \times \vec{S}^{(0)}(t) , \quad (98)$$

then reduces Eq. (88) to a scalar differential equation,

$$[\mathcal{D}^* - \frac{1}{4\tau_{\perp}(\varepsilon)}]\tau(\varepsilon) + \frac{1}{\tau_{IA}(\varepsilon)} = 0 , \quad (99)$$

which determines the generalized relaxation time  $\tau(\varepsilon)$ . Because the differential operator  $\mathcal{D}^*$  accounts for inelastic scattering, we have to conclude that even on the long time scale, where the spin polarization decays, inelasticity cannot be ignored. Thus, inelastic scattering processes not only determine the initial condition for the decay stage but they directly affect the time evolution (of the macroscopic spin polarization) in the decay stage. Multiplying from the left Eq. (99) by  $p(\varepsilon)\tau(\varepsilon)$ , integrating the resulting equation over  $\varepsilon$ , and using condition (84) yields an equivalent differential equation,

$$[\mathcal{D} - \frac{1}{4\tau_{\perp}(\varepsilon)}]p(\varepsilon)\tau(\varepsilon) + \frac{p(\varepsilon)}{\tau_{IA}(\varepsilon)} = 0 , \quad (100)$$

which can be also used to determine  $\tau(\varepsilon)$ .

Inserting Eq. (98) into (96), ignoring the  $\Omega_g$  term, and performing the angle and energy averages finally yields for the DP spin relaxation tensor

$$\mathbf{\Gamma}_{\text{DP}} = \frac{1}{\tau_{\text{DP}}} \begin{pmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 2 \end{pmatrix} , \quad (101)$$

with the DP spin relaxation rate given by

$$\frac{1}{\tau_{\text{DP}}} = \pi \langle \frac{\tau}{\tau_{IA}} \rangle_{\varepsilon} , \quad (102)$$

where the energy average is defined in Eq. (75). To determine the function  $p(\varepsilon)$ , we integrate Eq. (58), which gives

$$\left[ -v(\varepsilon) + \frac{d}{d\varepsilon} w(\varepsilon) \right] p(\varepsilon) = 0 , \quad (103)$$

where we used again condition (84) and introduced the dynamical friction and diffusion coefficients in  $\varepsilon$ -space,  $v(\varepsilon) = \varepsilon/\tau_f(\varepsilon)$  and  $w(\varepsilon) = \varepsilon^2/\tau_d(\varepsilon)$ , respectively. Integrating once more, we obtain

$$p(\varepsilon) = p(0) e^{\int_0^\varepsilon d\varepsilon' \frac{v(\varepsilon') - w'(\varepsilon')}{w(\varepsilon')}} , \quad (104)$$

with  $w'(\varepsilon) = dw(\varepsilon)/d\varepsilon$  and a normalization constant  $p(0)$ , which we fix according to

$$\int_0^{2\pi} d\phi \int_0^\infty d\varepsilon p(\varepsilon) = 1 . \quad (105)$$

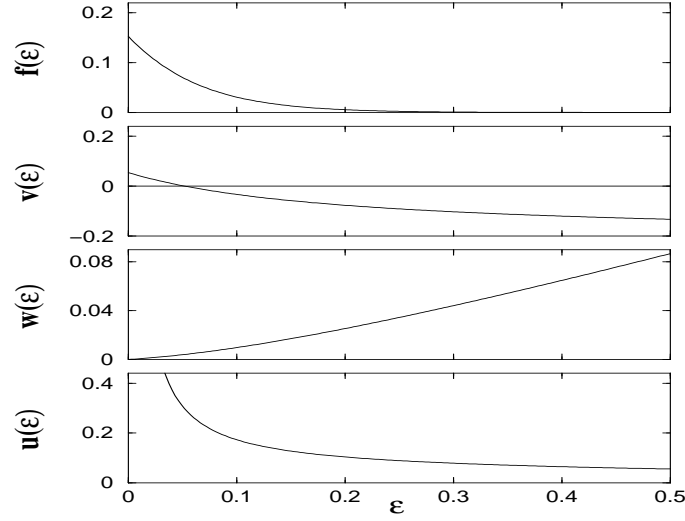


FIG. 3: Distribution function (for the equilibrated state at  $t \rightarrow \infty$ ), dynamical friction coefficient, dynamical diffusion coefficient, and angle randomization coefficient (from top to bottom) for a modulation doped quantum well at an electron density  $n = 4 \times 10^9 \text{ cm}^{-2}$  and  $T = 10 \text{ K}$ . For the calculation of the coefficients, only electron-electron scattering is taken into account. The unit of  $\varepsilon$  is  $14.93 \text{ meV}$ . The units of the functions  $v$ ,  $w$ , and  $u$ , are  $22.68 \text{ meV/ps}$ ,  $338.44 (\text{meV})^2/\text{ps}$ , and  $1.52 \text{ 1/ps}$ , respectively.

Note, because of condition (84), the integral in Eq. (104) is zero for  $\varepsilon \rightarrow 0$ . That is,  $p(\varepsilon)$  is well-defined for  $\varepsilon \rightarrow 0$ .

The Bloch equation (94) for the macroscopic spin polarization has to be solved with the spin relaxation tensor (101) and taking the particular geometry of the experimental set-up into account. Here, we consider the case of Kerr or Faraday rotation experiments, where the small magnetic field, which causes the spin precession, is along the x-axis. The propagation direction of the pump and probe pulses is assumed to be perpendicular to the quantum well plane, i.e., parallel to the z-axis (growth axis). The initial spin polarization is therefore along the z-axis, i.e.  $\vec{e} = (0, 0, 1)^T$ , and the probe pulse monitors the decay of a spin polarization which precesses in the yz-plane. Note that the spin decay in the yz-plane plane is not isotropic ( $\Gamma_{yy} \neq \Gamma_{zz}$ ). Assuming  $\tau_{\text{DP}} \gg 1/\Omega_L$ , the solution of the Bloch equation is

$$\delta \vec{S}(t) = \begin{pmatrix} 0 \\ -\sin \Omega_L t \\ \cos \Omega_L t \end{pmatrix} e^{-\Gamma t}, \quad (106)$$

where the decay rate of the spin polarization is given by the arithmetic mean of the decay rates in y- and in z-direction:  $\Gamma = (\Gamma_{yy} + \Gamma_{zz})/2 = 3/(2\tau_{\text{DP}})$ .

The results presented below are for a  $L = 25 \text{ nm}$  GaAs quantum well. The parameter needed to specify  $\hbar \vec{\Omega}_{IA}^{QW}(\vec{k})$  is  $\delta_0 = 0.06 \hbar^3 / \sqrt{(2m^*)^3 \epsilon_g}$ . [13] The remaining parameters, such as the effective CB electron mass or the static dielectric constant  $\epsilon_b$  (needed for the Coulomb matrix element) can be found in standard data bases.[64] Numerically, we first calculate  $v(\varepsilon) = \varepsilon/\tau_f(\varepsilon)$ ,  $w(\varepsilon) = \varepsilon^2/\tau_d(\varepsilon)$ , which define the differential operator  $\mathcal{D}$ , and the on-shell rate  $u(\varepsilon) = 1/4\tau_{\perp}(\varepsilon)$  taking electron-electron and electron-impurity scattering into account (see Appendix). We then determine  $p(\varepsilon)$  from Eqs. (104) and (105). Finally, we solve the differential equation (100) for  $\tau(\varepsilon)$  numerically and obtain the DP relaxation rate  $1/\tau_{\text{DP}}$  by numerically integrating Eq. (102).

In Figs. 3 and 4 we show, for  $T = 10 \text{ K}$ , the dimensionless functions  $v(\varepsilon) = \varepsilon/\tau_f(\varepsilon)$ ,  $w(\varepsilon) = \varepsilon^2/\tau_d(\varepsilon)$ , and  $u(\varepsilon) = 1/4\tau_{\perp}(\varepsilon)$  for a modulation doped quantum well with electron density  $n = 4 \times 10^9 \text{ cm}^{-2}$  (non-degenerate electrons) and  $n = 4 \times 10^{11} \text{ cm}^{-2}$  (degenerate electrons), respectively. In a modulation doped quantum well electron-impurity scattering is negligible because of the spatial separation between the dopants and the electrons, we take therefore only electron-electron scattering into account. The physical content of the functions  $v(\varepsilon)$  and  $w(\varepsilon)$  is that of dynamical friction and diffusion coefficients (in  $\varepsilon$ -space) for the “test” spin polarization resulting from the scattering between the spin-polarized “test” electrons and the equilibrated “field” electrons. The function  $u(\varepsilon)$  denotes the on-shell scattering rate arising from the “test” electron’s elastic scattering off “field” electrons. It randomizes the angle  $\phi$ ; hence  $u(\varepsilon)$  can be interpreted as an angle randomization coefficient.

In Figs. 3 and 4 we also show the electron distribution functions which characterize the equilibrated state at  $t \rightarrow \infty$ . Because of our assumption that only a small portion of the total number of electrons initially contributed to the “test” spin polarization (i.e.  $\delta f(\vec{k}, t) \ll f(\vec{k})$ ), the equilibrium distribution functions are used to determine

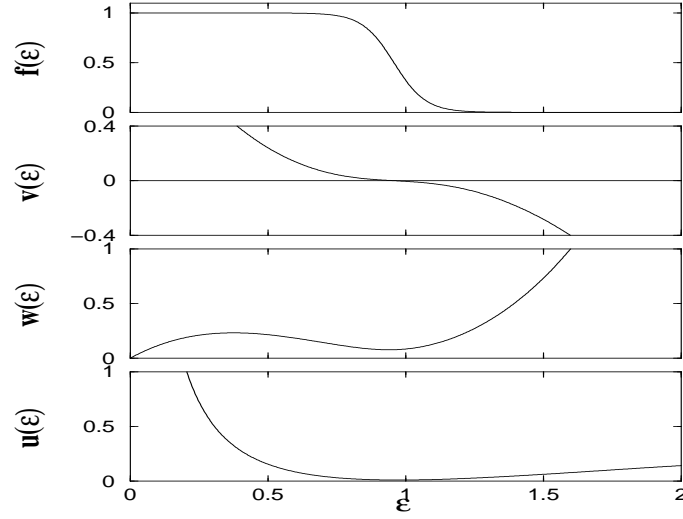


FIG. 4: Distribution function (for the equilibrated state at  $t \rightarrow \infty$ ), dynamical friction coefficient, dynamical diffusion coefficient, and angle randomization coefficient (from top to bottom) for a modulation doped quantum well at an electron density  $n = 4 \times 10^{11} \text{cm}^{-2}$  and  $T = 10 \text{K}$ . As in Fig. 3, for the calculation of the coefficients, only electron-electron scattering is considered. The units of  $\varepsilon$ ,  $v$ ,  $w$ , and  $u$  are the same as in Fig. 3.

the friction, diffusion, and angle randomization coefficients (see section III.B). In other words, we approximated at  $t < \infty$  the distributions of the friction and diffusion causing spin-balanced “field” electrons by the distributions of the equilibrated electrons at  $t \rightarrow \infty$ , despite the fact that at  $t < \infty$  of course a small number of electrons is still out of equilibrium and contributes to the finite spin polarization. Obviously, this is only permissible close enough to equilibrium.

Inelastic scattering encoded in  $v(\varepsilon) = \varepsilon/\tau_f(\varepsilon)$  and  $w(\varepsilon) = \varepsilon^2/\tau_d(\varepsilon)$  gives rise to spin-conserving energy relaxation and diffusion, which in turn yields a quasi-stationary spin polarization  $\delta \vec{S}_{st}(\varepsilon, \phi) = p(\varepsilon)\vec{e}$ , towards which any initial spin polarization relaxes very quickly without losing spins. The time scale on which this spin-conserving relaxation takes place is set by the relaxation times  $\tau_i(\varepsilon)$ ,  $i = f, d, \perp$ . In Figure 5 we show  $p(\varepsilon)$  for  $T = 10 \text{K}$  and three densities:  $n = 4 \times 10^9 \text{cm}^{-2}$ ,  $n = 4 \times 10^{11} \text{cm}^{-2}$ , and  $n = 4 \times 10^{12} \text{cm}^{-2}$ . In the inset we again depict the corresponding distribution functions for the electrons which characterize the equilibrated state at  $t \rightarrow \infty$ . At very low densities, where the electrons are non-degenerate,  $p(\varepsilon)$  is centered around  $\varepsilon = 0$ , while at high densities, where the electrons are degenerate,  $p(\varepsilon)$  is centered around the Fermi energy for the electrons. Note, however, that  $p(\varepsilon)$  describes the spin polarization at  $t = 0$  while  $f(\varepsilon)$  is the equilibrium distribution of the electrons at  $t \rightarrow \infty$ .

At this point, a brief discussion about the applicability of the diffusion approximation for the linearized collision integral  $J_{ee}^{(0)}$  is in order. The diffusion approximation is expected to be applicable because Eq. (37) has the form of a master equation and can thus be formally expanded with respect to the momentum transfer  $\vec{q}$ . For a sufficiently rapidly decaying transition probability  $W^{ee}(\vec{k}; \vec{q})$ , the expansion can then be truncated after the second order term as in Eq. (45). The validity of the diffusion approximation depends therefore on the transition probability, which in dimensionless form reads [see Eq. (A5) in the Appendix]

$$W^{ee}(\vec{k}; \vec{q}) = \int_{-\infty}^{\infty} d\omega |M(q)|^2 [f(k^2 - \omega) + n(-\omega)] \text{Im}\tilde{\chi}(q, \omega) \delta(\vec{k} \cdot \vec{q} - \frac{q^2}{2} - \frac{\omega}{2}), \quad (107)$$

with  $M(q) = 1/(q + q_s)$  the Coulomb matrix element,  $q_s$  the Thomas-Fermi screening wave number, and  $f(x)$  and  $n(x)$  the Fermi and Bose functions, respectively. Note, in Eq. (107) we do not expand the distribution functions and energies. As a result, cut-off problems are avoided and the friction and diffusion coefficients,  $A_i^{ee}(\vec{k})$  and  $B_{ij}^{ee}(\vec{k})$  respectively, can be calculated without restricting the  $\vec{q}$ -integration. Thus, the diffusion approximation does not ignore hard scattering processes with large momentum transfer; it only treats them approximately, whereas soft scattering processes are treated exactly. Moreover, from Eq. (107) it follows that  $J_{ee}^{(0)}$  is actually a “phonon-type” collision integral, the role of phonons being played by the collective excitations of the spin-balanced “field” electrons. Indeed, replacing  $M(q)$  by the electron-phonon matrix element and  $\text{Im}\tilde{\chi}(q, \omega)$  by the phonon spectral function, Eq. (107) gives the transition probability for spin-polarized “test” electrons scattering off equilibrated phonons. This analogy already suggests that a diffusion approximation is applicable to  $J_{ee}^{(0)}$ . To demonstrate its validity, it is however necessary to

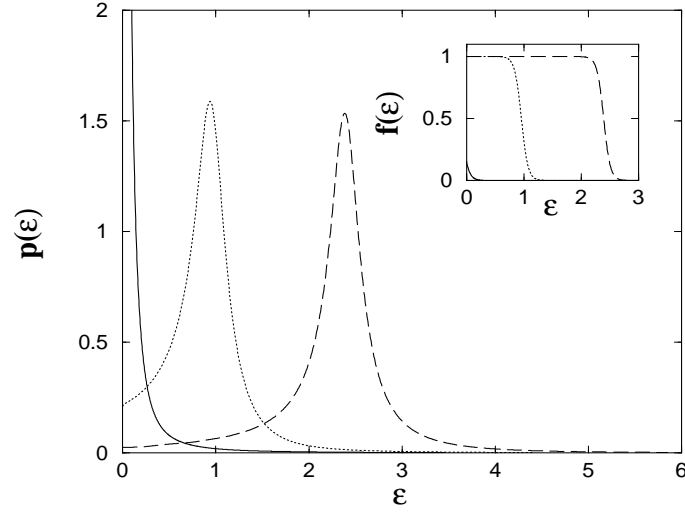


FIG. 5: Weight function for a modulation doped quantum well at  $T = 10\text{K}$  and three electron densities  $n = 4 \times 10^9 \text{cm}^{-2}$  (solid line),  $n = 4 \times 10^{11} \text{cm}^{-2}$  (dotted line),  $n = 10^{12} \text{cm}^{-2}$  (dashed line), taking only electron-electron scattering into account. The inset shows the corresponding electron distribution functions in the equilibrated state at  $t \rightarrow \infty$ .

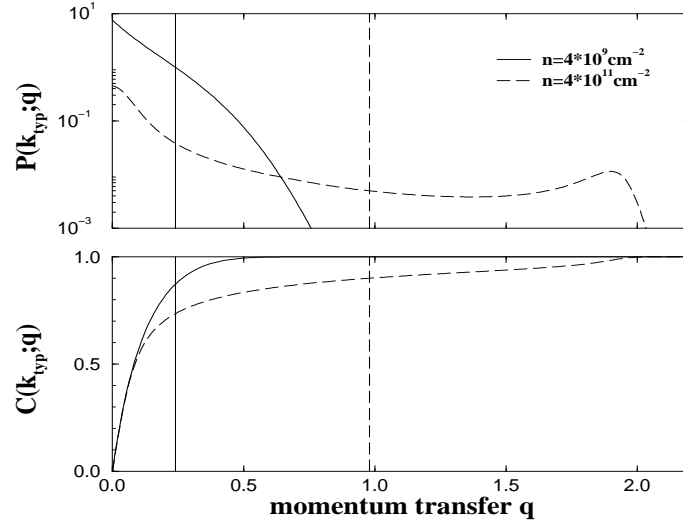


FIG. 6: The upper panel shows  $P(k_{\text{typ}}; q)$  for  $T=10\text{K}$  and two electron densities:  $n = 4 \times 10^9 \text{cm}^{-2}$  (non-degenerate electrons) and  $n = 4 \times 10^{11} \text{cm}^{-2}$  (degenerate electrons). For non-degenerate electrons, the typical momentum  $k_{\text{typ}}$  corresponds to the thermal energy (vertical solid line), whereas for degenerate electrons,  $k_{\text{typ}}$  is the Fermi wave number (vertical dashed line). In the lower panel we present the cumulant  $C(k_{\text{typ}}; q)$ , defined in Eq. (109), which is a measure of the relative importance of the scattering processes with a momentum transfer less than  $q$ .

show that for typical values of  $\vec{k}$  the transition probability  $W^{ee}(\vec{k}; \vec{q})$  indeed decays sufficiently rapidly for large  $\vec{q}$ .

For that purpose we investigate

$$P(k_{\text{typ}}; q) = \int d\phi \, q \, W^{ee}(\vec{k}_{\text{typ}}; \vec{q}) , \quad (108)$$

which is essentially the angle-averaged zeroth order moment of  $W^{ee}(\vec{k} = \vec{k}_{\text{typ}}; \vec{q})$  with  $\phi$  the angle between  $\vec{k}_{\text{typ}}$  and  $\vec{q}$ . [The first and second moments appear in the calculation of the friction and diffusion coefficients  $A_i^{ee}(\vec{k})$  and  $B_{ij}^{ee}(\vec{k})$ .] For degenerate electrons  $k_{\text{typ}}$  is the Fermi wave number  $k_F$  while for non-degenerate electrons  $k_{\text{typ}}$  is the wave number corresponding to the thermal energy. In Fig. 6 we show for the two parameter sets used in Figs. 3 and 4, respectively,

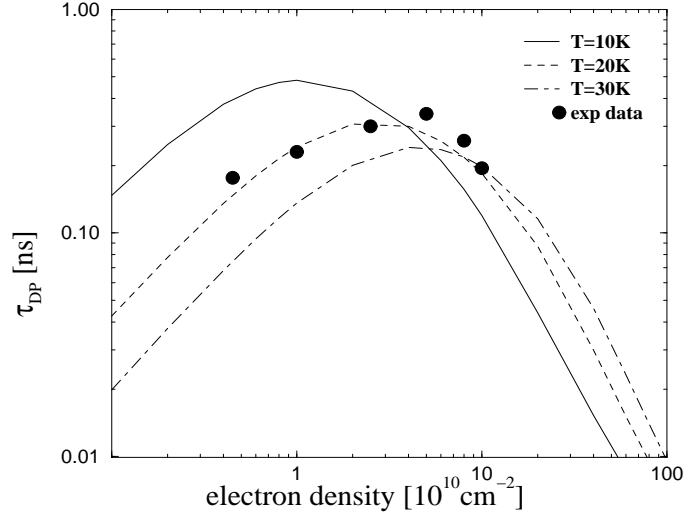


FIG. 7: D'yakonov-Perel' spin lifetime due to electron-electron scattering for a 25nm modulation doped quantum well as a function of electron density at three temperatures  $T = 10\text{K}$ ,  $T = 20\text{K}$ , and  $T = 30\text{K}$ . The solid dots are experimental data at  $T = 10\text{K}$  from Ref. [30].

the zeroth order moment  $P(k_{\text{typ}}; q)$  together with the cumulant

$$C(k_{\text{typ}}; q) = \frac{\int_0^q dr P(k_{\text{typ}}; r)}{\int_0^\infty dr P(k_{\text{typ}}; r)}, \quad (109)$$

from which we can estimate the relative importance of scattering processes with momentum transfer up to  $q$ . In the upper panel of Fig. 6 we see that  $P(k_{\text{typ}}; q)$  indeed decays with increasing  $q$ . In the degenerate regime  $q = 2k_F$  scattering is clearly visible, but one order of magnitude less probable than the  $q \rightarrow 0$  scattering process. From the cumulants, displayed in the lower panel, we infer moreover that in the non-degenerate as well as degenerate regime, soft scattering processes with  $q \leq k_{\text{typ}}$  give the main contribution (around 80-90 %). In both cases we expect therefore the diffusion approximation to produce reasonable quantitative results for the DP spin relaxation time. To estimate the error precisely is complicated. It would require a detailed investigation of the full momentum dependence of  $W^{ee}(\vec{k}; \vec{q})$  together with a reference calculation which does not invoke the diffusion approximation. The spin relaxation times we obtain as a function of temperature and density compare favorably with experimental results, indicating that the modeling of scattering processes within a diffusion approximation is sufficient for the calculation of DP spin relaxation times.

We now turn to the numerical results for the DP spin lifetime  $\tau_{DP}$ . Figure 7 shows  $\tau_{DP}$  as a function of electron density for a modulation doped quantum well at three temperatures  $T = 10\text{K}$ ,  $20\text{K}$ , and  $30\text{K}$ . Since in a modulation doped quantum well electron-impurity scattering is negligible, we take only electron-electron scattering into account. For fixed temperature, the spin lifetime first increases with electron density, reaches a maximum, and then decreases again. The non-monotonic density dependence of the spin lifetime follows the density dependence of the electron-electron scattering rate. At low densities, the scattering rate is small because of lack of scattering partners, while at high densities the scattering rate is suppressed because of efficient Pauli blocking. At intermediate densities, where the cross-over from non-degenerate to degenerate electrons occurs, the electron-electron scattering rate, and thus the DP spin lifetime, is maximal. The position of the maximum shifts with decreasing temperature to lower densities because the density, where the cross-over from a non-degenerate to a degenerate electron gas takes place, decreases with temperature. The relaxation time of photo-currents in optically pumped semiconductors shows a similar non-monotonic density dependence. [65]

For a fixed electron density, the DP spin lifetime decreases with temperature in the low density regime and increases with temperature in the high density regime. The latter is because of the temperature induced reduction of the Pauli blocking, giving rise to an increasing electron-electron scattering rate and therefore to an increasing DP spin lifetime. In the low density regime, on the other hand, increasing temperature broadens the electron distribution function, i.e. electrons occupy states higher up in the band. The average thermal energy increases therefore and the spin decay occurs preferentially from states higher up in the band, where the torque force induced by the bulk inversion asymmetry is larger. As a consequence, the DP spin lifetime decreases with temperature in the low density regime.

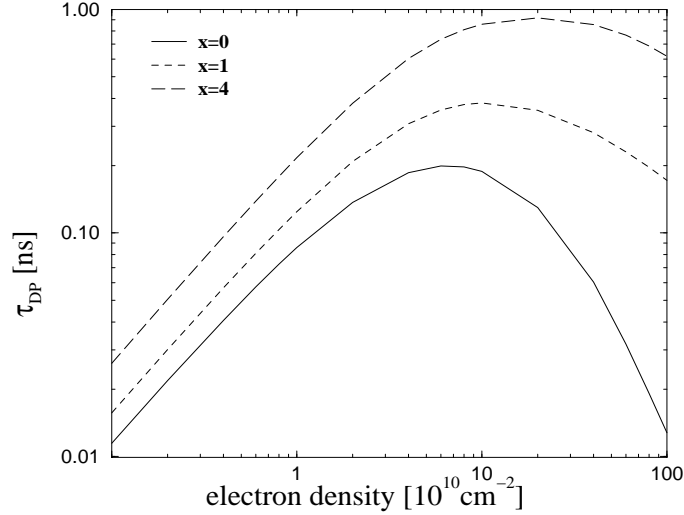


FIG. 8: D'yakonov-Perel' spin lifetime for a 25nm quantum well at  $T = 40\text{K}$  and three values of ionized (donor and acceptor) impurity concentrations: no impurities ( $x = 0$ ), ionized impurity concentration equal to the electron density ( $x = 1$ ), and ionized impurity concentration equal to four times the electron density ( $x = 4$ ). For  $x = 0$ , only electron-electron scattering contributes to the spin lifetime, whereas for  $x \neq 0$  both electron-electron and electron-impurity scattering determine the spin lifetime.

In Fig. 7 we also plot experimental data for  $T = 10\text{K}$  from Ref. [30]. For electron densities above  $n = 5 \times 10^{10}\text{cm}^{-2}$  the agreement between theory and experiment is quite good, given the fact that our calculation is based on an idealized quantum well with infinitely high confinement potential. In this density regime, we expect our results to even underestimate the spin lifetimes, because the model for the electronic structure of the quantum well most probably overestimates  $\hbar\tilde{\Omega}_{IA}^{QW}$ . Indeed, the constant defining the magnitude of the splitting of the conduction subband  $C_{IA}^{QW} \sim (\pi/L)^2 \sim E_1^\infty$ , with  $E_1^\infty$  the confinement energy of the lowest conduction subband. For a finite confinement potential,  $E_1$  is smaller than  $E_1^\infty = \hbar^2\pi^2/2m^*L^2$ , giving rise to a smaller splitting and, consequently, to a larger  $\tau_{DP}$ . To obtain in this density regime better agreement between experimentally measured and theoretically calculated spin lifetimes an improved electronic structure calculation is clearly necessary. At lower densities, on the other hand, electrons are most likely localized to donors (at  $T = 10\text{K}$  thermal ionization is negligibly small) and our theory, which is based on a band picture, does not apply.

In quantum wells which are not modulation doped, electron-impurity scattering due to donors and acceptors (in compensated samples) provides an additional, very efficient scattering process. The DP spin lifetime increases with scattering rate. As a result, we expect the spin lifetimes in quantum wells that are not modulation doped to be substantially longer than in modulation doped quantum wells. This can be seen in Fig. 8, where we plot the electron density dependence of the DP spin lifetime at  $T = 40\text{K}$  for  $x = 0$  (modulation doped),  $x = 1$  (uncompensated quantum well with equal impurity and electron density), and  $x = 4$  (compensated quantum well with impurity (donor and acceptor) density four times the electron density). As expected, the spin lifetimes increase with  $x$  for all electron densities. The increase is however not uniform, with the largest increase taking place at high electron densities, where Pauli blocking very effectively suppressed the DP spin lifetime in the modulation doped quantum well. The electron-impurity scattering rate is not affected by Pauli blocking and leads therefore to a substantial enhancement of the DP spin lifetime at high doping levels.

For a fixed density the character of the electron gas also changes with temperature. In particular, increasing temperature pushes the electron gas into the non-degenerate regime. For some temperature, the cross-over from a degenerate to a non-degenerate electron gas occurs, electron-electron scattering is particularly strong, and we expect spin lifetimes to be enhanced. [48] This effect is demonstrated in Fig. 9, where we plot for an electron density  $n = 3 \times 10^{10}\text{cm}^{-2}$  the temperature dependence of the spin lifetime for  $x = 0, 1$ , and  $4$ . In the modulation doped case ( $x = 0$ ) the enhancement of the spin lifetime in the temperature range where the cross-over from degenerate to non-degenerate takes place can be most clearly seen. For finite  $x$ , the spin lifetimes are for all temperatures longer than for  $x = 0$ . The enhancement is again not uniform. At high temperatures it is very small. While at low temperatures it is very large, because in that range the Pauli blocking leads to a strong suppression of the DP spin relaxation rate in modulation doped quantum wells. In fact, the spin lifetimes at low temperatures saturate by a value set by the electron-impurity scattering rate. The maximum in the spin lifetime is therefore less pronounced (or even disappears

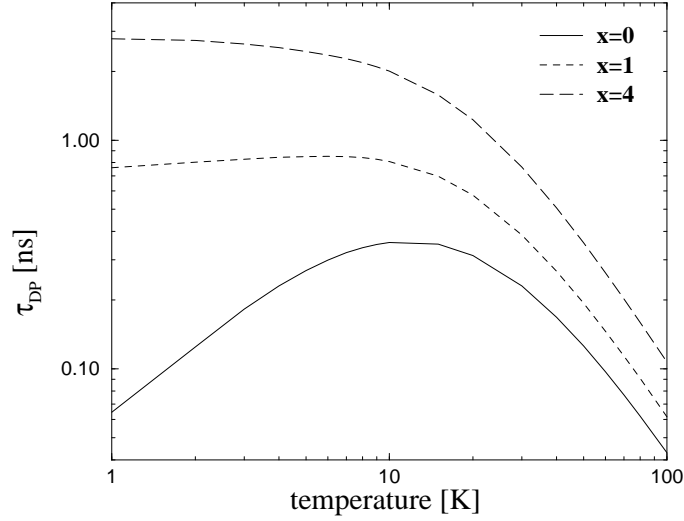


FIG. 9: D'yakonov-Perel' spin lifetime for a 25nm quantum well as a function of temperature at an electron density  $n = 3 \times 10^{10} \text{cm}^{-2}$  and three values of ionized (donor and acceptor) impurity concentrations: no impurities ( $x = 0$ ), ionized impurity concentration equal to the electron density ( $x = 1$ ), and ionized impurity concentration equal to four times the electron density ( $x = 4$ ). As in Fig. 8, for  $x = 0$ , only electron-electron scattering contributes to the spin lifetime, whereas for  $x \neq 0$  both electron-electron and electron-impurity scattering determine the spin lifetime.

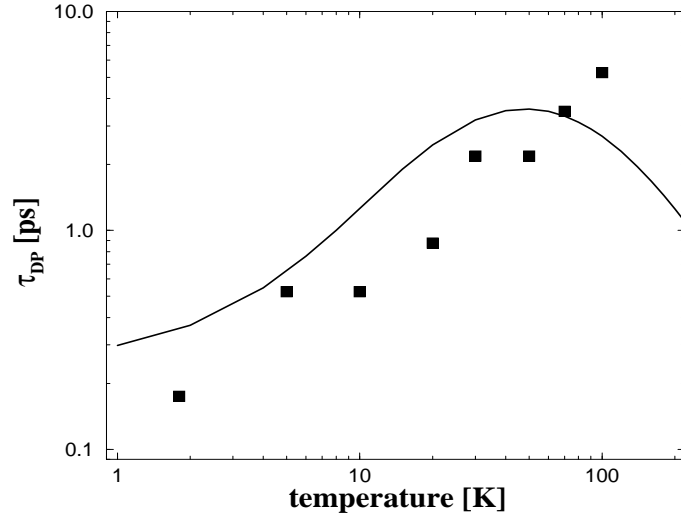


FIG. 10: D'yakonov-Perel' spin lifetime for a 10nm modulation doped GaAs quantum well as a function of temperature at an electron density  $n = 1.86 \times 10^{11} \text{cm}^{-2}$  (solid line). The solid boxes are experimental data from Ref. [47].

completely) in quantum wells that are not modulation doped.

In Fig. 10, we finally compare for a 10nm modulation doped GaAs quantum well with an electron density  $n = 1.86 \times 10^{11} \text{cm}^{-2}$  the theoretically obtained temperature dependence of the D'yakonov-Perel' spin lifetime with the experimentally measured temperature dependence of the D'yakonov-Perel' spin lifetime. [47] Below 50K the agreement between the experimental data points and the theoretical results is quite reasonable, suggesting that in this temperature range electron-electron scattering is the main source of electron spin relaxation in these samples. This conclusion is also supported by the theoretical results obtained by Glazov and coworkers. [48] In contrast to the theoretically predicted non-monotonic behavior with a maximum at  $T \approx 50\text{K}$ , the experimental results suggest that, for this electron density, the D'yakonov-Perel' spin lifetime  $\tau_{DP}$  grows monotonically with temperature. This is probably due to the neglect of phonons in the theoretical modeling. The maximum of  $\tau_{DP}$  is a consequence of electron-electron scattering. Additional scattering processes destroy the maximum. For instance, electron-impurity scattering increases the spin lifetimes at low temperatures, resulting in a monotonically decreasing spin lifetime (see  $x \neq 0$  curves in Fig.



9). The samples in Ref. [47] are high-quality quantum wells, where electron-impurity scattering should be negligible. The electron density  $n = 1.86 \times 10^{11} \text{cm}^{-2}$  is however rather high. As a result, the temperature, at which  $\tau_{DP}$  is expected to be maximal, falls in a temperature range, where electron-phonon scattering is significant. We expect a calculation, which takes electron-electron and electron-phonon scattering into account, to produce a monotonically increasing  $\tau_{DP}$ .

In this section we illustrated our semiclassical kinetic theory of spin relaxation by calculating the DP spin lifetime for an (idealized) quantum well, at temperatures and electron densities, where electron-electron and electron-impurity scattering dominate. Electron-electron scattering has been chosen to illustrate the effects of inelasticity and Pauli blocking. Electron spin lifetimes due to electron-electron scattering have been also calculated in Refs.[38, 44, 46, 48, 66], using however different approaches and focusing mostly on different aspects. In particular, the non-monotonous temperature and density dependence has not been addressed until quite recently. [47, 48] In modulation doped quantum wells, spin lifetimes turn out to be particularly long for electron densities and temperatures, where the cross-over from the non-degenerate to degenerate regime occurs. In this regime, many-body effects beyond the Born approximation are most probably important and should be included in a more quantitative calculation of the spin relaxation times. We expect however our main conclusions to be independent of the particular modeling of the Coulomb interaction.

## V. CONCLUSIONS

Starting from the full quantum kinetic equations for the electron Green functions we derived a (semiclassical) Fokker-Planck equation for the non-equilibrium spin polarization, assuming small spin polarizations and soft scattering. The Fokker-Planck equation conceptualizes the non-equilibrium spin dynamics in terms of a “test” spin polarization, comprising a small number of spin polarized “test” electrons, which scatter off an equilibrated bath consisting of impurities, phonons, and spin-balanced “field” electrons. Because of the scattering, the bath causes for the spin polarization dynamical friction, diffusion, and relaxation (decay). We then employed a multiple time scale perturbation approach to separate the fast spin-conserving from the slow spin non-conserving time evolution. As a result, we extracted from the Fokker-Planck equation a Bloch equation which controls the time evolution of the macroscopic ( $\vec{k}$ -averaged) spin polarization on the long time scale, where the spin polarization decays. Our semiclassical approach accounts for elastic and inelastic scattering and avoids the ad hoc energy averaging of on-shell spin relaxation rates. Instead we show that the weight function is intimately linked to the “quasi-stationary” spin polarization, which is the terminating state of the fast, spin-conserving time evolution taking place immediately after spin injection. The diagonal elements of the macroscopic ( $\vec{k}$ -averaged) spin relaxation tensor are the spin lifetimes. They are either given by an energy averaged spin-flip rate (EY process) or an energy average of a generalized relaxation time multiplied by a precession rate (DP and VG processes).

The formal development of our approach is based on a generic model for non-magnetic III-V semiconductors and treats EY and motional narrowing (DP and VG) spin relaxation processes on an equal footing. We also allowed for orbital motion of the electrons in a strong magnetic field, which potentially leads to a quenching of the motional narrowing-type spin relaxation processes. The derivation of the Fokker-Planck equation is independent of dimensionality and, as long as a soft scattering regime can be identified, also of the scattering processes, which enter the Fokker-Planck equation in the form of dynamical friction and diffusion coefficients, which have to be worked out separately for each scattering process.

To illustrate our formalism we applied it to a quantum well at low temperatures, where electron-electron and electron-impurity scattering dominate. We explicitly constructed the friction, diffusion and angle randomization coefficients characterizing the (symmetry-adapted) Fokker-Planck equation for that particular situation and calculated the DP spin lifetime at vanishingly small magnetic field as a function of electron density and temperature. We found that for fixed temperature (density) the density (temperature) dependence is non-monotonic. Spin lifetimes are particularly long for densities and temperatures, where the cross-over from a non-degenerate to a degenerate electron gas occurs. Spin lifetimes in compensated quantum wells are always longer than in modulation doped quantum wells with the same electron density. The enhancement of the spin lifetime is particularly strong for densities and temperatures where Pauli blocking is most efficient in suppressing the DP spin lifetime (due to electron-electron scattering) in modulation doped quantum wells.

Various extensions of our approach are conceivable and constitute research directions for the future. Semiconductor structures with structural inversion asymmetry [39] and/or native interface asymmetry [31] can be studied within our approach by augmenting the model Hamiltonian by the corresponding spin off-diagonal Hamiltonian matrix elements. In particular, the role of the linear collision integral  $J_B^{(1)}[f, \delta\vec{S}]$ , which does not affect spin lifetimes in isotropic semiconductors, should be reinvestigated, e.g., for an asymmetric quantum well where spin lifetimes can be particularly long because motional narrowing processes due to bulk and structural inversion asymmetry can be made to

cancel each other. [43, 49] Since  $J_B^{(1)}[f, \delta\vec{S}]$  potentially mixes spin relaxation channels, it could affect the cancellation. A Fokker-Planck equation of the form (27), perhaps augmented by additional driving terms, could be the starting point for a systematic calculation of spin transport coefficients (e.g., spin diffusion length) for spatially inhomogeneous systems, such as interfaces or biased heterostructures. Finally, non-linear effects due to large spin polarizations could be studied either at the level of the matrix-Boltzmann equation for the electronic density matrix [51, 52] or, if the diffusion approximation is used to simplify the collision terms, at the level of a “Fokker-Planck-Landau equation” for the spin polarization, where the differential operator describing spin-conserving scattering events as well as the spin-flip tensor explicitly depend on the spin polarization and the distribution of the spin polarized electrons.

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## APPENDIX A: CALCULATION OF RELAXATION RATES

In this appendix we calculate for a quantum well the dynamical friction and diffusion coefficients  $A_i(\vec{k})$  and  $B_{ij}(\vec{k})$  taking electron-electron and electron-impurity scattering into account. As a result, we obtain the relaxation rates  $1/\tau_f(\varepsilon)$ ,  $1/\tau_d(\varepsilon)$ , and  $1/\tau_\perp(\varepsilon)$  which define the differential operator  $\mathcal{D}(\vec{k})$  as well as the spin-flip tensor  $\mathbf{R}(\vec{k})$ . Since we discuss in this paper only the DP process quantitatively, which originates from the interplay of the momentum scattering encoded in the differential operator  $\mathcal{D}(\vec{k})$  and the torque force due to inversion asymmetry, it suffices to give explicit expressions only for the differential operator  $\mathcal{D}(\vec{k})$ . The derivation of the spin-flip tensor  $\mathbf{R}(\vec{k})$  proceeds along the same lines.

Within the diffusion approximation, the spin conserving collision integral  $J_\nu^{(0)}[f, \delta\vec{S}]$  becomes a Fokker-Planck differential operator (45) with dynamical friction and diffusion coefficients,  $A_i^\nu$  and  $B_{ij}^\nu$ , defined in Eqs. (46) and (47), respectively. First, we consider electron-electron scattering and calculate  $A_i^{ee}$  and  $B_{ij}^{ee}$ . To avoid the cut-off problem at large momentum transfers, which usually plagues the diffusion approximation to electron-electron scattering, we keep the full integrands in Eqs. (46) and (47), i.e., we do not expand the distribution functions and energies with respect to the momentum transfer  $\vec{q}$ . Using the identities  $f(\vec{k})f(\vec{k} \pm \vec{q}) = [f(\vec{k} \pm \vec{q}) - f(\vec{k})]n(\varepsilon(\vec{k}) - \varepsilon(\vec{k} \pm \vec{q}))$  and  $\delta(\varepsilon(\vec{k}) + \varepsilon(\vec{k}') - \varepsilon(\vec{k} - \vec{q}) - \varepsilon(\vec{k}' + \vec{q})) = \int d\omega \delta(\varepsilon(\vec{k}) - \varepsilon(\vec{k} - \vec{q}) - \omega) \cdot \delta(\varepsilon(\vec{k}') - \varepsilon(\vec{k}' + \vec{q}) + \omega)$ , we rewrite the transition probability (41) into

$$W^{ee}(\vec{k}; \vec{q}) = 2 \int_{-\infty}^{\infty} d\omega |V(q)|^2 \text{Im}\chi(q, \omega) [n(-\omega) + f(\vec{k} - \vec{q})] \delta(\varepsilon(\vec{k}) - \varepsilon(\vec{k} - \vec{q}) - \omega), \quad (\text{A1})$$

where we introduced the susceptibility of noninteracting electrons

$$\text{Im}\chi(q, \omega) = 2\pi \sum_{\vec{k}} [f(\vec{k} + \vec{q}) - f(\vec{k})] \delta(\varepsilon(\vec{k} + \vec{q}) - \varepsilon(\vec{k}) - \omega). \quad (\text{A2})$$

The Coulomb potential  $V(q)$  is taken to be statically screened with the screening length given by the Thomas-Fermi expression. Had we allowed for dynamical screening,  $V(q) \rightarrow V(q, \omega) = V_0(q)/\epsilon(q, \omega)$ . If  $\epsilon(q, \omega)$  is approximated by the RPA expression, the resulting Fokker-Planck equation would be at the level of a quantum analog to the Lenard-Balescu equation. [56] The calculation of the relaxation rates presented below could be also performed with this more general expression for the Coulomb matrix element. For simplicity we present here however only the results for the statically screened Coulomb potential.

To proceed, we introduce dimensionless quantities, measuring energies and lengths in scaled Rydbergs and Bohr radii, respectively. In particular, we use  $\tilde{R}_0 = R_0/s$ ,  $\tilde{a}_0 = \sqrt{s}a_0$ , with  $R_0\tilde{a}_0^2 = \hbar^2/2m_0$  and  $e^2 = 2\sqrt{s}\tilde{R}_0\tilde{a}_0$ , and choose  $s$  such that  $\tilde{R}_0 = 1\text{meV}$ . The dimensionless Fokker-Planck operator has the same form as in Eq. (45) with dynamical friction and diffusion coefficients given by

$$A_i^{ee}(\vec{k}) = C_{ee}^m \int d\vec{q} q_i W^{ee}(\vec{k}; \vec{q}), \quad (\text{A3})$$

$$B_{ij}^{ee}(\vec{k}) = \frac{C_{ee}^m}{2} \int d\vec{q} q_i q_j W^{ee}(\vec{k}; \vec{q}), \quad (\text{A4})$$

with  $C_{ee}^m = sm^*/\varepsilon_b^2\pi m_0$  and

$$W^{ee}(\vec{k}; \vec{q}) = \int_{-\infty}^{\infty} d\omega \frac{F(k^2, \omega, q)}{(q + q_s)^2} \delta(\vec{k} \cdot \vec{q} - \frac{q^2}{2} - \frac{\omega}{2}) , \quad (\text{A5})$$

where  $q_s$  is the Thomas-Fermi screening wave number and  $\varepsilon_b$  is the static dielectric constant. The function  $F(k^2, \omega, q)$  originates from the statistics of the electron gas and is given by

$$F(k^2, \omega, q) = \text{Im}\tilde{\chi}(q, \omega)N(k, \omega) , \quad (\text{A6})$$

$$\text{Im}\tilde{\chi}(q, \omega) = \int d\vec{k} [f(k^2 + \omega) - f(k^2)] \delta(\vec{k} \cdot \vec{q} + \frac{q^2}{2} - \frac{\omega}{2}) , \quad (\text{A7})$$

$$N(k, \omega) = f(k^2 - \omega) + n(-\omega) , \quad (\text{A8})$$

with  $f(x)$  and  $n(x)$  the Fermi and Bose functions, respectively. To calculate  $A_i^{ee}(\vec{k})$  and  $B_{ij}^{ee}(\vec{k})$  for a [001] quantum well, we first evaluate the integrals for a fixed coordinate system in which  $\vec{k} = k\hat{e}_x$  and then rotate to an arbitrary coordinate system. The result can be cast into the form

$$A_i^{ee}(\vec{k}) = -\frac{8}{k}G(k)k_i , \quad (\text{A9})$$

$$B_{ij}^{ee}(\vec{k}) = \frac{2}{k}H(k)\delta_{ij} + \frac{2}{k^3}E(k)k_ik_j , \quad (\text{A10})$$

respectively, with three functions defined by

$$G(k) = -\frac{C_{ee}^m}{4k} \int_{-\infty}^{k^2} d\omega \int_{q_{min}}^{q_{max}} dq \frac{qF(k^2, \omega, q)}{(q + q_s)^2} \frac{z}{\sqrt{1 - z^2}} , \quad (\text{A11})$$

$$H(k) = \frac{C_{ee}^m}{2} \int_{-\infty}^{k^2} d\omega \int_{q_{min}}^{q_{max}} dq \frac{q^2F(k^2, \omega, q)}{(q + q_s)^2} \sqrt{1 - z^2} , \quad (\text{A12})$$

$$E(k) = -\frac{C_{ee}^m}{2} \int_{-\infty}^{k^2} d\omega \int_{q_{min}}^{q_{max}} dq \frac{q^2F(k^2, \omega, q)}{(q + q_s)^2} \frac{1 - 2z^2}{\sqrt{1 - z^2}} . \quad (\text{A13})$$

Note that these integrals are well defined. The range of integration originates from the  $\phi$ -integration which also gives rise to the factor involving  $z = (q^2 + \omega)/2kq$ . With Eqs. (A9) and (A10) and a transformation to the radial variable  $\varepsilon = k^2$ , the spin-conserving part of the dimensionless electron-electron collision integral is given by (recall that in two dimensions  $\delta\vec{S}(\varepsilon, \phi, t)$  contains the factor  $J(\varepsilon)/(2\pi)^2n_s = 1/8\pi^2n_s$ )

$$J_{ee}^{(0)}[f, \delta\vec{S}] = \mathcal{D}_{ee}(\vec{k})\delta\vec{S}(\varepsilon, \phi, t) , \quad (\text{A14})$$

with

$$\mathcal{D}_{ee}(\vec{k}) = -\frac{\partial}{\partial\varepsilon}v_{ee}(\varepsilon) + \frac{\partial^2}{\partial\varepsilon^2}w_{ee}(\varepsilon) - u_{ee}(\varepsilon)\mathcal{L}^2 . \quad (\text{A15})$$

Here we have introduced the total angular momentum operator in two dimensions,  $\hat{\mathcal{L}} = -i\partial/\partial\phi$ , and the friction, diffusion and angle randomization coefficients,  $v_{ee}(\varepsilon)$ ,  $w_{ee}(\varepsilon)$ , and  $u_{ee}(\varepsilon)$ , which are linear combinations of the functions  $G(k)$ ,  $H(k)$ , and  $E(k)$  taken at  $k = \sqrt{\varepsilon}$ . Specifically, they read:

$$v_{ee}(\varepsilon) = -\frac{2C_{ee}^m}{\varepsilon^{1/2}} \int_{-\infty}^{\varepsilon} d\omega \int_{q_{min}}^{q_{max}} dq \frac{\omega F(\varepsilon, \omega, q)}{(q + q_s)^2} \frac{1}{\sqrt{1 - z^2}} , \quad (\text{A16})$$

$$w_{ee}(\varepsilon) = 4C_{ee}^m\sqrt{\varepsilon} \int_{-\infty}^{\varepsilon} d\omega \int_{q_{min}}^{q_{max}} dq \frac{q^2F(\varepsilon, \omega, q)}{(q + q_s)^2} \frac{z^2}{\sqrt{1 - z^2}} , \quad (\text{A17})$$

$$u_{ee}(\varepsilon) = \frac{C_{ee}^m}{\varepsilon^{3/2}} \int_{-\infty}^{\varepsilon} d\omega \int_{q_{min}}^{q_{max}} dq \frac{q^2F(\varepsilon, \omega, q)}{(q + q_s)^2} \sqrt{1 - z^2} . \quad (\text{A18})$$

The range of integration depends on the sign of  $\omega$ ,  $q_{max} = q_{max}^-(\varepsilon, \omega)\Theta(-\omega) + q_{max}^+(\varepsilon, \omega)\Theta(\omega)$  and  $q_{min} = q_{min}^-(\varepsilon, \omega)\Theta(-\omega) + q_{min}^+(\varepsilon, \omega)\Theta(\omega)$ , with

$$q_{min}^-(\varepsilon, \omega) = -\sqrt{\varepsilon} + \sqrt{\varepsilon - \omega} , \quad \omega \leq 0 , \quad (\text{A19})$$

$$q_{max}^-(\varepsilon, \omega) = \sqrt{\varepsilon} + \sqrt{\varepsilon - \omega} , \quad \omega \leq 0 , \quad (\text{A20})$$

$$q_{min}^+(\varepsilon, \omega) = \sqrt{\varepsilon} - \sqrt{\varepsilon - \omega} , \quad 0 \leq \omega \leq \varepsilon , \quad (\text{A21})$$

$$q_{max}^+(\varepsilon, \omega) = \sqrt{\varepsilon} + \sqrt{\varepsilon - \omega} , \quad 0 \leq \omega \leq \varepsilon . \quad (\text{A22})$$

The functions  $v_{ee}(\varepsilon)$  and  $w_{ee}(\varepsilon)$  are the dynamical friction and diffusion coefficients for the spin polarization in  $\varepsilon$ -space. They originate from the scattering of the “test” electrons comprising the spin polarization with the equilibrated, spin-balanced “field” electrons. Because the scattering is inelastic, the “test” electrons gain ( $\omega < 0$ ) or lose ( $\omega > 0$ ) energy by scattering off “field” electrons. The on-shell function  $u_{ee}(\varepsilon)$  describes randomization of the angle  $\phi$ . The integrals defining  $v_{ee}(\varepsilon)$ ,  $w_{ee}(\varepsilon)$ , and  $u_{ee}(\varepsilon)$  have to be done numerically. The singularities are integrable and Gaussian integration proved to be efficient. The limiting values are  $\lim_{\varepsilon \rightarrow 0} u_{ee}(\varepsilon) = u_0/\varepsilon$ ,  $\lim_{\varepsilon \rightarrow 0} w_{ee}(\varepsilon) = 0$ , and  $\lim_{\varepsilon \rightarrow 0} v_{ee}(\varepsilon) = v_0$ . Moreover,  $w_{ee}(\varepsilon) = v_0 \varepsilon$  for  $\varepsilon \rightarrow 0$ , i.e.  $dw_{ee}(0)/d\varepsilon = v_{ee}(0)$ , which is essential to guarantee spin conservation of the differential operator  $\mathcal{D}_{ee}(\vec{k})$  [cp. Eq. (84)]. The structure of the differential operator suggests to write  $v_{ee} = \varepsilon/\tau_f^{ee}$ ,  $w_{ee} = \varepsilon^2/\tau_d^{ee}$ , and  $u_{ee} = 1/4\tau_{\perp}^{ee}$ , with  $1/\tau_f^{ee}$ ,  $1/\tau_d^{ee}$ , and  $1/\tau_{\perp}^{ee}$  relaxation rates describing energy relaxation, diffusion, and randomization of the angle due electron-electron scattering, respectively.

The calculation of the (on-shell) relaxation rate due to electron-impurity scattering proceeds along the same lines. The starting point is Eq. (37), specialized to electron-impurity scattering, that is, with  $W^\nu(\vec{k}, \vec{q})$  given by Eq. (40). Going through the same steps as in the case of electron-electron scattering yields for the spin-conserving part of the dimensionless electron-impurity collision integral (as before  $\delta\vec{S}(\varepsilon, \phi, t)$  contains the factor  $1/8\pi^2 n_s$ )

$$J_{ei}^{(0)}[f, \delta\vec{S}] = \mathcal{D}_{ei}(\vec{k})\delta\vec{S}(\varepsilon, \phi, t), \quad (\text{A23})$$

where the differential operator is now given by

$$\mathcal{D}_{ei}(\vec{k}) = -u_{ei}(\varepsilon)\mathcal{L}^2, \quad (\text{A24})$$

with

$$u_{ei}(\varepsilon) = \frac{C_{ei}^m}{\varepsilon^{3/2}} \int_0^{2\sqrt{\varepsilon}} dq \frac{q^2}{(q+q_s)^2} \sqrt{1-z^2}, \quad (\text{A25})$$

$C_{ei}^m = s(4\pi m^* n_i \tilde{a}_0^2 / \varepsilon_b^2 m_0)$  and  $z = q/2\sqrt{\varepsilon}$ , where  $n_i$  is the sheet density of the impurities. Because of the elasticity of electron-impurity scattering, the differential operator contains only an on-shell term. The function  $u_{ei} = 1/4\tau_{\perp}^{ei}$  defines the relaxation time due to electron-impurity scattering. It only causes randomization of the angle  $\phi$ . The total spin-conserving collision integral, taking electron-electron and electron-impurity scattering into account, is given by  $D(\vec{k}) = D_{ee}(\vec{k}) + D_{ei}(\vec{k})$ .

Similar expressions can be derived for electron-phonon scattering. For bulk semiconductors the calculation proceeds along the same lines with the obvious modifications due to the additional angle integration.

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